Fluid Flow through Faults and Fractures in Argillaceous Formations

Proceedings of a Joint NEA/EC Workshop
Berne, Switzerland, 10-12 June, 1996

Organised by
the NEA Working Group on Measurement and Physical Understanding of Groundwater Flow through Argillaceous Media (the “Clay Club”)
and hosted by
the National Co-operative for the Disposal of Radioactive Waste, NAGRA (Switzerland)

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- assessing the contribution of nuclear power to the overall energy supply by keeping under review the technical and economic aspects of nuclear power growth and forecasting demand and supply for the different phases of the nuclear fuel cycle;
- developing exchanges of scientific and technical information particularly through participation in common services;
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FOREWORD

Many national programmes on radioactive waste management are considering geologic disposal in argillaceous formations. In order to determine the suitability of such media for waste disposal, evaluations of the transport of radionuclides from the disposal system to the accessible environment must be undertaken. These evaluations require not only site-specific data from a site characterisation programme, but also a sound general understanding of the basic physical and chemical processes that govern water, gas and solute transport through these formations. In this context, the NEA Co-ordinating Group on Site Evaluation and Design of Experiments for Radioactive Waste Disposal (SEDE) established the Working Group on Measurement and Physical Understanding of Groundwater Flow through Argillaceous Media (informally named the "Clay Club") to address the many issues associated with this subject. This Working Group promotes a continuing intercomparison of the properties of the different argillaceous media under consideration for geological disposal, and an exchange of technical and scientific information, by means of meetings, workshops and written overviews, on in situ characterisation of clays, and on water, gas and solute movement through these formations.

Groundwater, gas and solute will always tend to follow the paths of least resistance in their passage through low permeability formations. If such media are transected by permeable faults or interconnected fractures, cross-formational flow would be focused along these discontinuities. An evaluation of the occurrence of fluid flow through faults and fractures is thus of primary importance for the performance assessment of radioactive waste repositories located in argillaceous settings. Therefore, the Clay Club set up a workshop on this topic in order to provide the national waste management organisations and the scientific community at large with insights into the driving processes and the occurrence of fluid flow through faults and fractures in argillaceous formations.

As the European Commission has for many years promoted and co-funded, within the framework of its R&D programmes on radioactive waste management, research in the field of characterisation of argillaceous formations as potential media for geological repositories, it was agreed to have the workshop jointly organised by the Nuclear Energy Agency and the European Commission.

The workshop was hosted in Berne, Switzerland on 10-12 June 1996, by the National Co-operative for the Disposal of Radioactive Waste (NAGRA, Switzerland), who also lead a technical visit to the Mont Terri Underground Research Laboratory where in situ hydrogeological, geochemical and rock mechanical characterisation experiments are carried out in the Jurassic Opalinus Clay.

This publication includes the papers presented orally or as posters at the workshop, and is introduced by a synthesis of the topics addressed and the conclusions reached. The opinions expressed are those of the authors only, and do not necessarily reflect the views of any OECD Member country or international organisation. The proceedings are published on the responsibility of the Secretary-General of the OECD.
ACKNOWLEDGEMENTS

On behalf of all participants, the NEA and the EC wish to express their gratitude to the National Co-operative for the Disposal of Radioactive Waste (NAGRA, Switzerland) for its kind hospitality which undoubtedly contributed to the success of the workshop. They also wish to thank the members of the Programme Committee for the help provided in setting up and holding the workshop:

Marc Thury (NAGRA, Switzerland, Clay Club Chairman), Jean-François Aranyossy (ANDRA, France), Eric Frank (HSK, Switzerland), Andreas Gautschi (NAGRA, Switzerland), Ferruccio Gera (ISMES, Italy), Bert Haijting (EC), Steve Horsemann (BGS, United Kingdom) and Philippe Lalieux (OECD/NEA).

The NEA, the EC and the Programme Committee members are also very grateful to the speakers and the posters’ authors, and to all the workshop attendees for their active and constructive participation.

Philippe Lalieux, from the Radiation Protection and Waste Management Division of the OECD/NEA, is responsible for the scientific secretariat of the Clay Club and the organisation of this workshop.
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SYNTHESIS OF THE WORKSHOP
Fluid Flow through Faults and Fractures in Argillaceous Formations –
Synthesis of the Workshop

Philippe Lalieux
OECD/Nuclear Energy Agency

Steve Horsemann
British Geological Survey (United Kingdom)

1. INTRODUCTION

Groundwater, gas and solute will always tend to follow the paths of least resistance in their passage through low permeability formations. If such media are transected by permeable faults or interconnected fractures, cross-formational flow would be focused along these discontinuities. An evaluation of the occurrence of fluid flow through faults and fractures is thus of primary importance for the performance assessment of radioactive waste repositories located in argillaceous settings. Such an evaluation should also include several factors (e.g. plastic nature of argillaceous media, burial depth and history, fluid-rock interactions, swelling) that might tend to close, at least partially, faults and fractures and impede flow through these. Moreover, the specificities of the various argillaceous media under consideration (plastic clays, claystones, shales) and the timeframe upon which migration is considered should also be taken into account.

To provide national waste management organisations and the scientific community at large with insights into the driving processes and the occurrence of fluid flow through faults and fractures in argillaceous formations, the NEA Working Group on Measurement and Physical Understanding of Groundwater Flow through Argillaceous Media (informally named the “Clay Club”) set up a workshop on this subject. This workshop was jointly organised with the European Commission, and hosted in Berne, Switzerland, on 10-12 June 1996, by the National Co-operative for the Disposal of Radioactive Waste (NAGRA, Switzerland).

2. WORKSHOP OBJECTIVES AND SCOPE

The workshop aimed at the following objectives:

- to review and discuss state of the art understanding and methodologies addressing the issues of fluid flow and transport through faults in argillaceous media and their transferability to specific sites;
- to discuss the relevance of flow and transport through faults and fractures for repository safety and performance, and the need for characterising such processes; and
• to contribute to a better understanding of argillaceous media.

The workshop was introduced by a general paper on the relevance of fracture flow in argillaceous media to radioactive waste isolation. This presentation also detailed a series of key questions to be addressed in the course of the workshop that was elaborated by the Programme Committee:

• What are the predominant mechanisms for fluid flow and solute transport? What is the role of faults, joints, and microfractures? Are there preferential flow paths (channels) within faults?
• What are the predominant factors responsible for the increased permeability of these features?
• How can we distinguish between permanent (steady), time-limited and short-term episodic flow in these features?
• What are the predominant factors for self-healing of preferential flow paths?
• What kind of investigations are recommended to answer the above mentioned questions?
• What can be learned by experimental work? Do we get meaningful quantitative data? Do we increase process understanding?
• What can radioactive waste, hazardous waste and petroleum industry learn from each other concerning flow through fractures and faults in argillaceous media? Which findings are transferable (analogous)?

The first session provided the audience with a general overview on some of the basic concepts controlling the evolution of argillaceous rocks, and with a keynote presentation on the relevance of faults and fractures to fluid flow in these geologic settings. The other sessions addressed the driving processes of flow through faults and fractures and presented several case studies. A poster session dealt with additional case studies and with more specific, technical details. The outcomes of the oral and poster presentations provided the basis for the concluding discussion.

The workshop was based on the results and the needs of the national radioactive waste disposal programmes but, given the obvious relevance of experience from other applications, multidisciplinary approaches were solicited. In recognition of the substantial contribution of the Oil and Gas Industry to current understanding of fault sealing and fluid migration in compact mudrocks, it was an aim of the Programme Committee to attract participation from consultants and researchers with experience in these areas: approximately 50% of the presentations related to national waste disposal programmes, 30% to academic work, and 20% to oil and gas exploration and production. We are confident that these Proceedings adequately demonstrate the mutual benefits of this interchange. The workshop was attended by 76 delegates from 10 countries.

2. SCOPE OF THE SYNTHESIS

The present paper aims at (i) synthesising the key factors which control fracture flow; (ii) integrating the various scales of interest; and (iii) putting them in the perspective of the long-term performance of deep repositories located in argillaceous media. In preparing this synthesis, we felt that the presentations and published papers contained a number of very real insights into the role of faults and fractures in fluid flow in argillaceous rocks (i.e. mudrocks), with some degree of commonality of opinion. We have therefore opted to extract key passages from each paper and to
present them in a fairly logical order, with added commentary, so that the reader can form an independent opinion on these complicated issues.

The very wide spectrum of argillaceous media (from plastic overconsolidated clays with a high water content to highly compacted shales with a very limited water content), timeframes (minute observations in underground facilities to fluid migration over several hundreds of million years), spatial scales (microscopic analysis of fault gauge to regional basin studies), depths (sub-surface to several thousands of meters) and processes (rock-water interactions at the particle level to regional flow gradient) which were covered during the two-day presentations should be emphasised and borne in mind when assessing the role of the various processes, e.g. most of the case studies and laboratory experiments related to oil and gas migration and entrapment deal with hard, compacted shales at great depths (i.e. important pressure and temperature) and with very long timeframe. Table 1 provides a simplified overview of the main characteristics (objectives, geological settings, scales, and key conclusions) of the case studies presented at the workshop, and is aimed at replacing the statements quoted in this synthesis in their contexts. The overview papers’ which were presented during the first session, are excluded from the table according to the fact that they are not based on a few, specific cases but well represent synthesis of years of experience.

3. KEY FACTORS CONTROLLING FLUID FLOW THROUGH FAULTS AND FRACTURES

One of the primary attributes of argillaceous rocks is their low permeability when contrasted with most other sedimentary rocks. Thick sequences of clay-rich sediments are considered to act as aquitards, in so far as they generally impede the advective motion of groundwater during its passage through the geological system from the region of recharge to the region of discharge. In clays with a more compact fabric, orientation and overlapping of platy clay particles often leads to an additional reduction of the interconnection of pores in the direction normal to bedding. The very narrow channels between the clay particles may be extremely tortuous and the water molecules are therefore obliged to move in regions which are very close to mineral surfaces. This proximity of the mobile water to the mineral surfaces, together with the strength of water-clay interactions, is probably the principal reason for the low permeability of compact clay-rich media. Matrix permeability usually decreases very substantially as mudrock is subject to increasing compaction, far more than we would normally anticipate from the decrease in total porosity.

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1. Papers by A. Thury; A. Matter; A.G. Koestler and A.G. Milnes; and J.M. Logan. It is to note that the content of the presentation on “Influence of Shale on Fault Behaviour during Hydrocarbon Migration, Accumulation and Protection” made by A.G. Koestler has been integrated in the overview paper by A.G. Koestler and A.G. Milnes.
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<th>SCALE (DEPTH/THICKNESS)</th>
<th>KEY FEATURES / PROCESSES TIMEFRAME</th>
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<td>Muir-Wood (II)</td>
<td>Observations of hydrological response to active tectonics and of other transient hydrogeologic behaviours.</td>
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<tr>
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<td>Effects of stress, stress history and current depth of burial on pathway flow (critical state soil mechanics).</td>
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<td>Local to regional.</td>
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</tr>
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<td>Generic deep sand/shale layered complexes.</td>
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</tr>
<tr>
<td>Leythaeuser (II)</td>
<td>Recognition and quantification of the geochemical effects of expulsion of petroleum fluids from source rocks.</td>
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<td>Sample.</td>
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1 (P) = Poster Session  
2 d = depth, t = thickness
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<tbody>
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<td>Kenter et al. (II)*</td>
<td>Review of available numerical tools and operational experience for slurry reinjection in deep shales and liquid natural gas storage in unlined caverns.</td>
<td>Several case studies.</td>
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</tr>
<tr>
<td>Clennell et al. (II)</td>
<td>Microstructural and petrophysical analysis of faults and fault damage zones.</td>
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</tr>
<tr>
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<td>Establishing the efficiency of shales as seals to hydrocarbon gas diffusion and multiphase fluid flow (input data for basin modelling)</td>
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* No paper or extended abstract provided.
Table 1: (Continued from previous page)

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<td>Neuzil et al. (III)</td>
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</tr>
<tr>
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<td>Direct and indirect evidences of secondary permeability of clays.</td>
<td>Plio-Pleistocene Blue Clays; Cretaceous to Oligocene scaly, indurated Clays (Argille Scagliose); Miocene Laga Flysch; Oligo-to Miocene Numidian Flysch; Miocene Clays and Marls; (Italy and Sicily).</td>
<td>Tunnels, quarries and dams. (d=100s m)</td>
<td>Variable behaviour of gas and water flows in similar materials. Difficulties in assessing the secondary permeability at depth without extensive investigations.</td>
</tr>
<tr>
<td>Grainger et al. (III)</td>
<td>Use of the upward migration of natural gases as gas-permeable fault tracers.</td>
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</tr>
<tr>
<td>Koide et al. (III)</td>
<td>Qualitative evaluation of the hydraulic conductivity of a reverse fault zone by long-term head observations prior, during and after a shaft excavation.</td>
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<td>The fault acts as a low conductivity feature. Hydraulic conductivity of the fault zone is heterogeneous and linked with the thickness of the clay filling.</td>
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A. Discontinuities in sedimentary rocks

Matter (Session I) makes the introductory comments: “During shallow burial, porewaters are expelled from compacting argillaceous muds through the interparticle pore network. With increasing overburden and degree of particle orientation, accompanied by loss of porosity and permeability, flow is channelled along inhomogeneities such as sand layers, fractures and faults.”

On the subject of classification, Koestler and Milnes (Session I) observe that discontinuities in sedimentary rocks fall into two main categories: primary – formed during deposition; and secondary – formed after deposition, during or after lithification. They add: “Tectonic fractures may be roughly subdivided in three main categories - joints, faults and veins/stylolites - depending on the observed movement of the fracture walls. Joints of presumed tectonic origin are ubiquitous in apparently undeformed, sub-horizontal, sedimentary sequences, but their genesis is problematic and controversial. They are generally sub-vertical, often bedding-confined, and usually arranged in joint domains of different or systematically changing preferred orientation, showing little relation to large-scale structures. Well-developed joint sets often show different characteristics in different lithological units, apparently related to the different mechanical properties of the rock-types and to variations in bed thickness. Joint surfaces show typical associations of plumose markings, conchoidal rib marks and fringes of en échelon minor joints, which can be interpreted in terms of the established principles of fracture mechanics.” The above features are typical of extensional (Mode I) fractures, with very small opening displacements perpendicular to the fracture surface. “In contrast to joints, faults are more localised and irregular, both in orientation and distribution, and the faulting process physically changes the rock properties along, and in the vicinity of the fault. It is convenient to distinguish different scales of structure: regional faults (major structures, with displacements measured in kilometres upwards), large faults (displacement of 10s to 100s of metres, visible on seismic reflection profiles), small faults (displacement of centimetres to metres, visible at outcrop but not in seismic sections) and microfaults (displacement of millimetres, visible in hand-specimen, core, or thin-section).”

B. Fault structures and the role of the damage zone

Koestler and Milnes state: “The zonation of bulk rock properties associated with a single large fault can thus be described in terms of the fault zone itself (breccia, gouge, clay smear), the damage and/or drag zone (which extends beyond the fault tip lines) and the surrounding undeformed rock. In well-bedded rocks and shale-dominated successions, ductile deformation and flexural-slip reduce the need for fracturing by producing a drag zone.” Logan (Session I) states: “The fault rock has been deformed by a variety of mechanisms depending on temperature and pressure regimes. These range from mechanical comminution, smearing of more ductile material, dislocation glide and creep, recrystallisation, to solution transport. Laboratory experimental work on simulated gouge has established a characteristic fabric that has been verified in many natural faults.” Koestler and Milnes observe: “The fringes around faults are more complex and variable than joint fringes, often associated with en échelon systems of tension gashes, where the concentrated shear displacement on the fault plane is translated into more distributed shear in the tip region, or with asymmetrical zones of extension and shortening as the displacement decreases. To a first approximation, small faults and microfaults can be thought of as forming an interconnected network in a zone adjacent to a large fault - the damage zone”. In a similar vein, Clennell et al. (Session II) comment: “Faults are rarely simple planes of slip between two domains of undeformed rock. Rather slip is more usually accommodated in a fault zone consisting of anastomosing and splaying slip planes. Faults are commonly surrounded
by a region of influence, where stresses associated with propagation and movement of the fault have produced a greater density of small faults and fractures than are found further away in the undeformed rock mass. Major and minor slip surfaces may be filled with cataclastic gouge, smeared clay or precipitated minerals. Overall displacement is partitioned between the large increments of slip on the main fault planes and smaller displacements on splays and minor faults.” Logan suggests that: “Flow parallel to the fault is facilitated in the damage zone, but flow at high angles to the fault is inhibited by the fine grain-size of the fault rock.”

Koestler and Milnes make the concluding remarks: “The degree of pore-closure and dewatering and the depth of burial determine the mechanical properties of clay-rich rocks in a complicated way which leads in some circumstances to ductile/plastic deformation (décollement, diapirism, smearing along faults) and in others to brittle deformation (hydrofracture, jointing in unconsolidated clay). Clay smearing along discontinuities significantly changes hydraulic properties of rock zones for both cross-flow and along-flow, mainly in a reducing way, but also in a flow increasing way especially when clay reacts in a brittle manner during reactivation and uplift of former deeply buried ductile sediments. These, however, are special effects which will be the main focus of the workshop.”

C. Field evidence of secondary (fracture) permeability

It has been surmised that the presence of pathways (faults and/or interconnected fractures) might explain a disparity between laboratory-determined permeabilities and those that must be assigned to a mudrock formation to explain regional groundwater flow (i.e. the scale-dependence of mudrock permeability). In invoking this scale-dependence, it seems that we must consider the degree of induration (diagenetic alteration) of the mudrock, the current depth of burial, the 3-D state of stress, and the stress history of the formation (i.e. overconsolidation and/or post-glacial compaction effects). Muir-Wood (Session II) comments: “The presence of potentially conductive apertures passing through low permeability formations reveals the difficulty of attempting to sample and scale the permeability of a formation from local observations.” Matter expresses the opinion: “Fractures are the single most important conduits in tight shales and largely control cross-formational flow”.

There is ample evidence that faults and fractures in de-stressed and highly-overconsolidated clays (close to surface) can be transmissive. In reference to the overconsolidated Oxford Clay of the UK (around 50m below ground surface), Metcalfe et al. (Poster Session) state: “The overall model is that modern meteoric water penetrates downwards through the mudrocks over the entire site, diluting the pre-existing groundwaters and porewaters. The fault acts to short-circuit groundwater flow and flushing has been particularly vigorous along this structure.” Walraevens et al. (Poster Session) examine a semi-confined Eocene aquifer in Flanders which is recharged in areas of higher topography where it is overlain by the Bartonian clay. Flow modelling of the recharge area, where piezometers provide detailed knowledge of the hydraulic heads, has indicated a vertical hydraulic conductivity for the clay of 10⁻⁷ m.s⁻¹. The comment is made: “Laboratory measurements often provide values which are at least one order of magnitude lower. This discrepancy can be ascribed to the presence of preferential pathways in the clay.” The authors present geochemical evidence for almost complete flushing of the original marine porewater. The base of the dipping clay stratum is from 20 to 30 m below surface in the recharge area.

With reference to the IPSN studies in the Toarcian claystones at the Tournemire (Aveyron, France) tunnel (around 230 m below ground surface, with exploratory boreholes above and below this
level), Boisson et al. (Session II) report that: “in the worst case, the increase in permeability values that might be ascribed to the presence of fractures would be about one order of magnitude $10^{13}$ to $10^{16}$ m.s$^{-1}$ for the fractured zones, and $10^{14}$ to $10^{15}$ m.s$^{-1}$ for the clayey matrix itself. This does not reveal any really significant differences.” Stable isotope measurements reported by Moreau-le Golvan et al. (Poster Session) on Toarcian claystone porewaters from samples taken from an unfractured zone indicate the presence of meteoric water, possibly recharged under cooler climatic conditions. Concerning the water in the fractured zone, the authors note: “That the porewater of the fractured zone could have the same meteoric origin, but would have been affected by a secondary process of enrichment in heavy isotope, which is not fully understood for the time being.”

Neuzil and Belitz (Session III) summarise studies of groundwater movement in the Cretaceous shales of North America which act as confining layers for underlying aquifers. According to these authors, the intact (i.e. matrix) permeability of the shales is quite consistent everywhere at $10^{-21}$ to $10^{-20}$ m$^2$ ($10^{-14}$ to $10^{-13}$ m.s$^{-1}$). The Hayes study area showed no evidence of secondary (i.e. fracture) permeability. The shale is underpressed and analysis of the subnormal pressures using a transient flow analysis yields in situ values for the shale permeability of $10^{-21}$ to $10^{-20}$ m$^2$, in good agreement with intact rock values. The absence of horizontal hydraulic gradients suggests that transmissive fractures cannot be present or are very widely separated, since they would tend to produce fairly rapid dissipation of the subnormal (disequilibrium) pressures. In the Denver Basin the situation is much the same and the range of values obtained by steady-state modelling agrees remarkably well with laboratory and in situ permeability values for the intact shale, and thereby indicates an absence of any secondary permeability. The authors comment: “This is a provocative result because it shows no permeability increase with scale in the shales over a surprisingly large area.” Calculation of hydraulic conductivities by the inverse analysis of anomalous pressures is also the subject of the paper by Vasseur et al. (Session II), but these authors focus on the phenomenon of overpressuring. They conclude: “Fluid overpressure is a common hydrodynamic phenomenon in sedimentary basins. Its occurrence mainly stems from the increasing gravity or tectonic loading under burial and from the resulting compaction of shale layers. Other causes such as diagenesis may also play some role. In any case, the generation and maintaining of overpressures over long geological periods imply that the medium is very tight.”

Gautschi (Session III) states: “Hydraulic tests in deeper boreholes, where the Opalinus Clay (Middle Jurassic shale) was encountered at depths between 300 and 1100 m, yielded very low hydraulic conductivities ($<10^{-12}$ m.s$^{-1}$), even though joints and faults were in some of the test intervals. Despite the complex tectonics of the folded Jura, comprising numerous reverse (thrust) and normal faults in a water-saturated environment, only two indications of humid patches (one in a fault containing calcite veins, one of unknown origin) have been reported from a total of 6400 m of tunnel sections in the Opalinus Clay. The only measurable water inflow reported was in connection with an intercalated calcareous sandstone layer, which is only developed in the north-western edge of Switzerland.”

Neuzil and Belitz also discuss analyses of regional flow in the Dakota Aquifer of N. and S. Dakota which provide very contrasting results to those mentioned above; these studies indicate much higher regional permeabilities for the Cretaceous shales. The authors observe: “Numerical simulation is only possible if the shales are assigned permeabilities in the range $10^{-16}$ to $10^{-18}$ m$^2$, which are from 2 to 5 orders of magnitude larger than intact rock permeabilities and indicative of secondary (fracture) permeability. We hypothesise that such transmissive fractures control the regional vertical permeability of (and leakage through) the shales, but only at depths less than approximately 1 kilometre. The fractures are very sparse and separated by a kilometre or more.” However, “It is
possible that there are no transmissive fractures in the shale at all”. Indeed, no such fractures have been observed and their occurrence is postulated based on indirect evidence (geochemical, geothermal and geomorphological) only. With reference to the same data set, **Horseman and Harrington** (Session II) observe: “Extrapolation of the hydraulic conductivity depth-trends for the overconsolidated Cretaceous shales of S. Dakota leads to the observation that the trend for local-scale (i.e. matrix) hydraulic conductivity converges with the regional trend at a critical depth of around 2 kilometres. This suggests that the fractures responsible for scale-dependency would be closed at this depth. We propose a possible relationship between this critical depth and the critical state (of theoretical soil mechanics) and note that a depth-related transition in material behaviour might explain the depth-trends in hydraulic conductivity observed in these materials.” **Mazurek et al.** (Poster Session) describe the investigation of the Valanginian Marl (Palfris Formation) at the NAGRA Wellenberg site (Helvetic nappes of the central Swiss Alps) and make the observations: “While the frequency and the geometry of faults do not vary significantly with depth, their transmissivity decreases substantially. The enhanced fracture apertures at shallow depths are also indicated by much lower shear-wave velocities when compared to the deeper parts of the boreholes.”

**D. Mechanics of fracturing and dilatancy in mudrocks**

In attempting to explain the postulated depth- and stress-dependency of fault/fracture flow in mudrocks, attention is drawn to the mechanics of fracturing. **Célérier et al.** (Poster Session) in their ongoing study on the impact of irreversible deformation on the hydraulic properties of clays make the following very important statement: “Three geological situations can lead to a stress path pointing towards the dilatant part of the yield envelope of a compacting rock: (1) the hydraulic connection of the compacting rock to an overpressured lower reservoir, (2) an erosion event which affects the vertical stress magnitude, and (3), a change in the tectonic regime that reduces at least one of the horizontal stress magnitudes. The behaviour of clays is different when they are subjected to extensional (\(\sigma_1 < \sigma_2 = \sigma_3\)) or compressional (\(\sigma_1 > \sigma_2 = \sigma_3\)) triaxial stress paths.”

When we couple these criteria with the statement by **Clennell et al.**: “Faults in clay and shale formations typically have a more compact fabric than the host sediments, so it is difficult to explain how they can behave as conduits for fluid flow unless they are dilated by some mechanism”, we arrive at a fairly comprehensive picture of the events which might lead to significant fault/fracture permeability in clay-rich media. **Clennell et al.** comment: “… even un lithified clays can behave in a macroscopically brittle fashion (and dilate) if they are subject to shear strain under conditions of effective normal stress significantly lower than the maximum previously attained.”

Drawing on critical state theory, **Horseman and Harrington** observe: “If significant fluid flow in a compact mudrock requires the development of dilated pathways, then it is seems clear that an overconsolidated mudrock on the “dry side” of critical will be more susceptible to the development of such pathways than a similar mudrock lying on the “wet side””. **Célérier et al.** make the unequivocal statement: “In other words, to obtain dilatancy the clay must be overconsolidated.” The role of dilatancy is also emphasised by **Thompson** (Poster Session) who notes: “The Rusey breccia (Cornwall, UK) was formed under unusually dilatant conditions, generating a zone of high porosity and permeability”. **Labaume et al.** (Poster Session) state: “The formation of the mineral veins is indicative of dilatancy in a direction opposite to the direction of compression responsible for the formation of the foliation.”
Overpressuring represents the first of the mechanisms identified by Céléri er et al. and we will refer to it as criterion (1). In reference to fluid flow in the Barbados accretionary prism, Dewhurst et al. (Poster Session) summarise their observations as follows: “It seems therefore likely that fracture pathways must be open sub-parallel to the décollement as a consequence of elevated fluid pressures to account for the high permeabilities observed in the natural system.” Horseman and Harrington note: “Under special circumstances, it would appear to be the case that very high fluid pressures can actually create the pathways of fluid migration through mudrock formations.” Céléri er et al. comment: “Natural fluid overpressures occurring in fine-grained sediments can be released by two different mechanisms: large scale natural hydrofracturing and smaller scale damage associated with dilatancy which enhances permeability.” Henriet and De Batist (Poster Session) discuss evidence for a major hydraulic fracturing and de-watering event in the Ieper (London) Clay of Belgium. Leythaeuser (Session II) notes the smaller-scale effects of overpressuring: “It is shown under which circumstances development of pore fluid pressures can lead to microfracturing of the rock matrix.”

Horseman and Harrington state: “The processes of uplift, erosion and exhumation of a mudrock, leading to overconsolidation and associated stress-relief, are evidently very important considerations when examining the propensity for fluid flow along preferential pathways in such materials.” They add: “The state of stress developed in a highly-overconsolidated mudrock at shallow depths can therefore lead to spontaneous shear failure and the formation of small reverse faults.” In relation to the Valanginian marl, Mazurek et al. make the observation: “The larger transmissivities at shallow depth are best explained by an increase of the fracture apertures and of the connectivity due to the decrease of overburden by erosion and/or post-glacial decompression.” These are both clearly references to criterion (2), above.

Koestler and Milnes observe: “Fracturing in little disturbed sedimentary rocks is often determined by the action of small, horizontal extensional strains, both on a large scale (normal faulting) and on a small-scale (extensional jointing and microfaulting).” Since horizontal extensile strains are associated with a change in tectonic stresses, this correspond with criterion (3), above, for dilatancy. Krooss et al. (Session II) outline a programme of work aimed at relating hydraulic conducivity in the damage zone of a fault to the tectonic stress field. For a sample exhibiting seismic anisotropy (> 4500 m.s⁻¹ in NNW-SSE direction, < 3500 m.s⁻¹ in ENE-WSW direction), a strong permeability anisotropy was established with 2 x 10⁻² m² in the high velocity direction and approximately 2 x 10⁻² m² in the low velocity direction. In relation to hydraulic tests in the Boom Clay (Tertiary) of Belgium, Ortiz et al. (Session III) make the comment: “It is clear that fluid flow is strongly influenced by local geomechanical conditions. A decrease in effective stress will lead to an increase in hydraulic conductivity.” Figure 7 of Krooss et al. also illustrates the relationship between permeability and effective stress in argillaceous rocks.

E. Fault sealing and sealing mechanisms

Logan observes: “The occurrence of faults that act as seals to fluid flow across them has become a common feature of many hydrocarbon basins and is now well documented by pressure and in some cases geochemical differences between fluid compartments.” Clovell et al. state: “Study of numerous smaller deformation features recovered from North Sea reservoirs shows, that the amount of slip, beyond a few centimetres, is relatively unimportant in forming a sealing lithology, provided that the temperature and stress are sufficient for a range of deformation mechanisms to operate.”
Clennell et al. also comment: "They (faults) are also normally considered to be self-sealing, because the ductile nature of clay enables the walls to slide past one another without generating asperities and open fractures. This is something of a misconception, because shales at depth can become brittle as they lithify, and so produce zones of fracturing that can increase overall permeability by many orders of magnitude." Horseman and Harrington provide an alternative point of view: "In most rocks, the brittle-ductile transition occurs at high levels of confining pressure and at the elevated temperatures typical of moderately deep crustal conditions. Mudrocks are exceptional in their behaviour and true plasticity can usually be attained at earth surface temperatures and relatively modest levels of effective confining pressure. In an overconsolidated mudrock, the brittle-ductile transition manifests itself in triaxial experiments performed in the laboratory as the gradual disappearance of the post-peak strain-softening response with increasing effective (confining) stress and an associated change from a highly-localised shear deformation to a more (macroscopically) homogeneous form of deformation." The capacity of deeply-buried shales to respond to gravitational instability by plastic deformation so as to form enormous diapiric structures and the very low shearing resistance of shales evident in décollements and similar thrust structures, each provide very strong indications of the central role of interparticle water in rheological behaviour, even in deeper buried and lithified clay sediments. However, Logan warns us not to place too much reliance on conventional (Terzaghi) concepts of effective stress: "In contrast to most other rocks, over a wide range of strain rates, shales do not behave in accordance to the concept of effective stress."

Clennell et al. observe: "Shear zones may have a permeability that is typically one or two orders of magnitude lower than that of the undeformed wall rock. Permeability reduction is caused by (i) increased particle alignment that increases tortuosity, (ii) the blocking an collapse of sheltered pores, and (iii) an overall reduction in porosity as a result of shear-induced consolidation. This type of seal is probably of primary importance in argillaceous formations, particularly at shallower levels." Logan states: "Fracture porosity and resulting permeability are highest in the damage zone, higher than in the relatively unfractured country rock adjacent to it; fluid flow parallel to the fault is preferentially enhanced in this domain." Clennell et al. examine other potential sealing mechanisms: "Cataclasis and post-deformation pressure solution may contribute significantly towards the porosity collapse within phyllosilicate-framework fault rocks. In argillaceous formations, cementation may be focused along more permeable, coarse-grained horizons, and in the vicinity of faults and fractures. Mineral precipitation may completely occlude fluid pathways. Thus, depending on their nature and location, cements can have a positive or negative effect on seal integrity."

Other potential sealing mechanisms include replacement of the macroporosity by a matrix of fine-grained phyllosilicates (either detrital or authigenic).

Dewhurst et al. describe laboratory measurements of permeability during shearing of clay from the décollement of the Barbados accretionary prism and observe: "Although the shear-parallel permeability is greater (up to 35 times) than the shear-normal permeability, it is of similar magnitude to that of the consolidated sediment. Weak dilation and strain softening were evident in the shear strength response of the sample, but neither the void ratio nor the permeability increased above that recorded during elastic rebound of the consolidated clay. The permeability measured along the décollement during packer tests is far higher than the shear-parallel intergranular permeability recorded in the experiments." Comparing the Barbados situation with a structure in the Pyrenean mountains, Labaume et al. comment: "In both cases, the clay shear fabric formation is associated with strong porosity collapse. However, different features (carbonate veins in a carbonate-free host-sediment in Barbados, the release of potassium during mica-dickite transformation and a second generation of calcite veinlets in the Pyrenean mountains) indicate open systems with preferential fluid flow circulation through the fault zones during or after deformation. Periods of high pore pressure
favoured dilation and formation of the veins and periods of low pore pressure and tectonic displacement lead to the shear bands."

Logan observes: "In clay-rich materials, the potential for faults to act as seals to fluid migration either across them, or along them, is much greater than in other rock types. The fault zone produces a condition of flow anisotropy." The medium-term hydrogeological impacts of the Tsukiyoshi fault at the PNC-operated Tono Mine (Japan) are illustrated by Koide et al. Before shaft excavation, drawdown caused by the existing gallery was observed on the upthrow side of the fault, while on the downthrow side the piezometric profile was hydrostatic. Furthermore, the transient behaviour of the piezometric head on the upthrow side could be correlated with the shaft excavation schedule, while the decay of piezometric heads on the downthrow side was more gradual. The fault zone consists of two clay layers only 2 to 3 cm thick, with 10 to 20 cm of unconsolidated sand. The measurements indicate that the permeability across the fault must be lower than that of the mixed lithology country-rocks.

F. Gases and hydrocarbons

Shales play an important role in hydrocarbon exploration and production because of their capability to seal faults and caprocks over long geological periods of time, to an extend which allows accumulation of hydrocarbon. In an overview paper, Koestler and Milnes presented data from the oil industry and discussed the sealing potential of faults in argillaceous media. Impey et al. (Poster Session) introduce the problem of gas transport in mudrocks: "There is however some debate about the validity of using continuum Darcy models to simulate gas migration in very low permeability media, such as the argillaceous formations considered for radioactive waste disposal. This is because, in such media, gas will tend to migrate along a small number of preferential pathways (paths of least resistance), which is not consistent with a continuum interpretation." Grainger et al. (Session III) conclude: "The main results of the gas leakage study by soil-gas surveys showed that leakage of terrestrial gas occurs in correspondence with tectonic fracture zones, both along the margin and within the centre of clay basins. This observation means that in spite of the great thickness and plasticity of the clay, if fractured it does not always form an impermeable barrier to naturally migrating gas; clays can prevent gas from rising only far away from highly fractured zones." Ortiz et al. observe: "Gas breakthrough is associated with the creation of preferential pathways. A non-Darcy behaviour of gas flow rate versus pressure was observed. No major desaturation resulted from the breakthrough."

Returning to the problem of shale seals, Krooss et al. state: "Although conceptually well-established, the quantification and prediction of capillary sealing efficiency constitutes one of the major problems in the appreciation of hydrocarbon seals. For oil or gas leakage through seals, the capillary forces must be overcome. Some of the most eminent open questions with respect to the capillary efficiency are the breakthrough conditions (breakthrough pressure, percolation threshold) for tight rocks." Measurement of the methane diffusivity of confined specimens of pelitic rocks (Jurassic shales, Carboniferous and Permian claystones) at 150°C gave values in the range 10⁻⁸ to 10⁻¹⁰ m².s⁻¹. A correlation was found between effective diffusivity and TOC. Hanebeck et al. (Poster Session) make the statement: "...it can be concluded that kerogene in organic matter rich source rocks is load-bearing. Kerogene conversion can cause fluid flow in the rock or overpressuring." Leythauser discusses evidence for the preferential expulsion of lower molecular weight hydrocarbons from the shale interval. The results are indicative of transport of the light fraction in gaseous solution.
Black et al. (Poster Session) shed some light on the problem of slug test deconvolution when the presence of gas is suspected.

G. Dynamic aspects of fluid flow

Logan makes the observation: “Seals are rarely static in time and space with cycles of rupturing followed by healing having been documented.” Clennell et al. also mention the fault-valve mechanism of episodic flow: “The shale was considered to be compliant enough to keep existing fractures and faults sealed at near-hydrostatic pressures. As overpressure builds (to super hydrostatic but sub-lithostatic fluid pressures) the stress differential is sufficient to initiate and hold open sub-vertical hydraulic fractures. The walls stay apart for some time (10’s to 100’s of years) permitting rapid fluid flow before the overpressure drops below the threshold necessary to keep the conduits open.” Horseman and Harrington observe that: “The basic requirements for the occurrence of this intermittent or episodic flow process may be summarised as: (a) a nonlinearity and/or threshold in the flow law, (b) partial or complete resealing of the flow pathway under a suitable stress state, (c) a source reservoir capable of recharging (i.e. repressurising with time), and (d) sufficient energy stored in the source reservoir to support the flow during active periods. Since a compressed gas stores substantially more energy than a liquid, gas-dominated systems are likely to be much more prone to episodic flow than liquid-dominated systems.” Labaume et al. also discuss variations in fault permeability with time and comment: “Quantification of these variations is an important goal for assessment of fault transport capacities and needs a better knowledge of controlling factors, principally the kinetics of fault-sealing mechanisms.” Muir-Wood addresses the seismic pumping mechanism and observes: “Seismic strain-cycling has the potential to change the hydrogeology of low permeability argillaceous materials, first by affecting the aperture of microcracks distributed through the formation and second by imposing altered fluid pressures at the boundaries, or within, the formation.”

H. Importance of faults/fractures in deep geologic disposal

We must be very wary of drawing any conclusions about the tightness of a formation from observation made at outcrop or at very shallow depths, since stress-relief and weathering are likely be a dominant factors affecting fault/fracture transmissivity in these “exhumed” and highly-overconsolidated mudrocks. Likewise, we must be cautious about making too strong inferences on the sealing capacity of argillaceous rocks from Oil and Gas Industry experience, because the deformation mechanisms operating in argillaceous rocks at typical source-rock depths may differ quite considerably from those taking place at repository depths. However, it is perfectly valid to consider properties and responses over a wide depth-range in the effort to establish and understand important trends with increasing depth, temperature and stress. On an unrelated issue, we observe that evidence of palaeocirculation cannot be construed as evidence of present-day groundwater circulation. On the basis of our current understanding of underpressuring in argillaceous rocks, the occurrence of this disequilibrium state would seem to provide fairly compelling evidence of minimal secondary permeability at a site. Because of the associated problem of natural hydrofracturing, overpressuring is a contra-indication for repository siting, but is not known or proven to occur in the depth-range and geological settings suitable for repository development.

Gera (Session III) concludes: “Despite the limitations due to the uncertainty of many data, the picture of a complex and difficult-to-predict behaviour is confirmed. Due to the many factors controlling the physical and hydraulic properties of argillaceous sediments, it is practically impossible
to predict the secondary permeability at depth without extensive site-specific investigations." Although we would be wise to heed this warning, it does seem unduly pessimistic since it suggests that we are unable to understand the relationship of faults and fractures to the geological history of any formation and cannot hope to comprehend the physical and chemical interactions which determine the transmissivity of these features.

Neuzil and Belitz make the summary comments: "In either case, the shale section is an island of relatively low permeability in mid N. America and demonstrates that heterogeneity of the shallow crust occurs at scales up to thousands of kilometres. The vast bulk of the shale appears to be controlled by the intact shale permeability, which is quite small. As a result, chemical mass within the shale is probably sequestered for geologically significant periods of time. This has important implications for the isolation of toxic materials."

Lancelot et al. (Session III), in their isotope study of the Marcoule silty-clay series (MSS) (Gard, France), state: "Rb/Sr, U/Pb and Pb/Pb data are consistent with an evolution of the isotopic tracers in closed system conditions since 100 Ma, without exchange of radiogenic or radioactive isotopes by water-rock interaction, in spite of the onset of the Pyrenean and Alpine orogenies, the Oligocene rifting and the Messinian event. In the Pb/Pb isochron diagram, Pb isotope evolution suggests a crystallisation age of 89 Ma for the authigenic pyrites disseminated in the MSS and a 42 Ma age for the fluid palaeocirculation in the fault (i.e. at the end of the Pyrenean compressional event or at the beginning of the Oligocene rifting)."

Lege and Shao (Poster Session) comment: "The conclusion is drawn that matrix diffusion is an important property of fractured claystone for contaminant retardation. This, as well as the hydraulic properties of the geologic barrier, has to be taken into account when a permanent repository for radioactive waste is assessed."

Gautschi notes: "The hydrogeological dataset presented in this paper, part of which is for a worst-case type geological environment (i.e. tectonically strongly disturbed), suggests that radionuclide transport through faults and joints is probably not a critical issue for assessing the suitability of Opalinus Clay as a host rock for a deep geological repository for nuclear waste."

Clellnell et al. observe: "We believe that fault zones in uncemented argillaceous formation probably have sufficient ductile compliance to remain intact, with the failure planes mated together, unless they are reactivated by earth movements or hydrofractured by elevated pore fluid pressures." They add: "Fault zones have been considered as possible loci for fluid flow. Experimental evidence suggests that the fabrics developed within such fault zones are not conducive to enhanced transport unless they become strongly dilated, or are embrittled and then breached by fractures."

4. CONCLUSIONS

This workshop was the second of its type to be organised by the Clay Club, and, like its predecessor, was distinguished by the quality of its presentations, the truly international flavour of the meeting, and the friendly and free exchange of ideas on matters of central importance to the understanding of argillaceous media and to the deep geologic disposal of radioactive wastes. The

interest of exchanging information with the Oil and Gas Industry, especially concerning a) geoscientific methodologies and approaches; b) analogies; and c) integration of all geoscientific data/approaches at different scales, was clearly demonstrated.

The main conclusion which emerged from the workshop is as follows: there are strong mechanical and chemical reasons for faults to be tight at repository depth (very good examples of real tightness and fault-sealing were presented even in difficult geological conditions); however, fluid flow through faults and fractures in argillaceous media cannot be excluded per se and may appear under certain particular conditions (e.g. overpressurisation at great depth or, potentially, in the presence of gas pressure build-up).

A general guideline for the siting of potential radioactive waste repositories in argillaceous media would be a site selection process targeted at stable sedimentary basins. This will provide additional confidence that the extent of fracturing is limited and that the prevailing stress regime does not favour dilated faults and/or fractures with significant aperture.

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WORKSHOP PROCEEDINGS
SESSION I

General Overview
Chairmen: F. Gera (ISMES, Italy), and B. Haisjink (EC)
Relevance for Waste Isolation of Flow through Faults And Fractures In Argillaceous Formations – Key Questions to Address during the Workshop

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Abstract

After a brief description of repository concepts and safety barrier systems for high-level radioactive waste disposal, some general considerations are made on groundwater flow and radionuclide transport through argillaceous formations as well as on escape from a repository of gases produced in situ. The paper concludes with a list of key questions to be addressed by the presentations and posters of the workshop.
Introduction

Many national programmes in radioactive waste management are considering geological disposal in argillaceous media. In order to determine their suitability for waste disposal, it is necessary to undertake evaluations of the transport of radionuclides from such a disposal system to the biosphere. These evaluations require not only site-specific data from a site-characterisation programme, but also a sound general understanding of the basic physical and chemical processes that govern solute transport through these formations.

Groundwater, gas and solute will always tend to follow the paths of least resistance in their passage through low permeability formations. If such media are transected by permeable faults or interconnected fractures, flow would be focused along these discontinuities. An evaluation of the occurrence of fluid flow through faults and fractures is thus of primary importance for assessment of radionuclide transport from radioactive waste repositories located in argillaceous media. Such an evaluation should also include several factors (e.g. the plastic nature of argillaceous media, the burial depth and history, the fluid-rock interactions, swelling) that might tend to close, at least partially, faults and fractures and impede flow through these. Moreover, the specific properties of the various argillaceous media under consideration (from plastic over-consolidated clays with a high water content to highly compacted shales with a very limited water content) and the time frame upon which migration is considered should also be taken into account.

Repository concepts and safety barrier systems

In many national programmes, repository concepts for the final disposal of vitrified high-level waste and spent fuel have been developed and published. They all foresee a series of disposal galleries in low permeability areas or blocks at a depth of a few hundred to 1000 m below surface, with access by shafts or ramps.

Two types of disposal systems have been proposed for a mined repository in argillaceous host rocks: a) disposal within the repository tunnel, and b) disposal in emplacement holes, drilled vertically or at an angle into the host rock beneath the repository tunnel.

Safety is based on the principle of multiple barriers provided e.g. by the waste form (borosilicate glass), engineered structures (canister, backfill) and the geological environment. It is expected that most radionuclides would decay to insignificant levels within the engineered barriers. The geological barriers provide a stable and protective environment for the engineered barriers, ensuring their longevity; they also have the potential to provide retardation (with consequent radioactive decay) of any radionuclides that eventually escape from the engineered barriers. Their nuclide retention capacity is optimised through the siting of the repository in a low-permeability host rock with favourable groundwater chemistry in a tectonically stable location. The series of multiple barriers for a high-level waste disposal system is shown schematically in Fig. 1. For long-lived intermediate-level wastes where disposal concepts in large diameter silos or galleries are foreseen, significant amounts of cementitious material may be used for waste conditioning, repository backfill or mechanical support.
### Safety barrier system for high-level waste

**Glass matrix (in steel mould)**
- Low corrosion rate of glass
- High resistance to radiation damage
- Homogeneous radionuclide distribution

**Spent fuel elements**
- Low UCs dissolution rate
- High radiological / thermal stability of UCs-matrix

**Steel canister**
- Completely isolates waste for > 1000 years
- Corrosion products act as a chemical buffer
- Corrosion products take up radionuclides

**Bentonite backfill**
- Long resaturation time
- Low solute transfer rate (diffusion)
- Retardation of radionuclide transport (sorption)
- Chemical buffer
- Low radionuclide solubility in leachate
- Colloid filter
- Plasticity (self-healing following physical disturbance)

**Geological barriers**
- Repository zone:
  - Low water flux
  - Favourable geochemistry
  - Mechanical stability

**Geosphere:**
- Retardation of radionuclides (sorption, matrix diffusion)
- Reduction of radionuclide concentration (dilution, radioactive decay)
- Physical protection of the engineered barriers (e.g. from glacial erosion)

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Figure 1. The safety barrier system for high-level waste (by courtesy of Nagra)
Groundwater flow and radionuclide transport

For the assessment of the radionuclide transport through argillaceous formations, the way of groundwater flow is of primary importance. Three different typical possibilities can be distinguished:

- Groundwater flow through sparsely spaced faults
- Groundwater flow through an interconnected fracture system (e.g. joints, bedding planes) and sedimentary layers of enhanced permeability (e.g. carbonate-rich or sandy layers)
- Quasi-stagnant groundwater (advective flow negligible, radionuclide transport by diffusion only).

The radionuclide retention of the rock is significantly different in these three cases:

- In the case of groundwater flow through single faults, radionuclides are transported with the groundwater through these faults, partly sorbed on the rock surfaces of the flow paths in these faults and partly fixed when they diffuse into the pores of the rock matrix along the faults.
- In the case of groundwater movement through a fracture system, the radionuclides are transported and retarded as in the case of faults, however, the rock mass accessible for diffusion is significantly bigger and therefore retardation is also significantly higher.
- In the case of stagnant groundwater, radionuclides can move through the rock pores by diffusion only, which is an extremely slow process. In addition, they can be significantly sorbed on the surfaces of clay minerals in the rock.

The last case described is by far the most favourable case with the strongest radionuclide retention in the rock. Therefore, it is of primary importance to evaluate the potential for groundwater flow through faults and fractures in argillaceous formations.

Repository gas release

In a deep geological radioactive waste repository, hydrogen gas may be generated in significant quantities by anaerobic corrosion of metallic canisters and (to a lesser extent) by radiolysis. In the case of permeable faults or fracture systems, this gas will escape through these features, which represent the paths of least resistance.

In the case of absence of permeable faults or fracture systems, the gas pressure may rise in the repository and lead to induced fracture systems propagating from the repository to the next permeable zone, e.g. the next aquifer. The key question is, if these induced fractures represent new permeable flow paths and lead to a significant reduction of the radionuclide retention.

Experience from hydrocarbon exploration (gas migration through argillaceous caprocks, caprock failure) might be helpful for the assessment of processes related to repository gas releases.

Key questions to be addressed during the workshop

A list of key questions to be addressed by the presentations and posters was elaborated by the Workshop Programme Committee. These questions should be related to the various types of argillaceous rocks under consideration (plastic clays, clay stones, shales):
• What are the predominant mechanisms (conceptual models) for fluid flow and solute transport? What is the role of faults, joints, and micro-fractures? Are there preferential flow paths (channels) within faults?

• What are the predominant factors responsible for the increased permeability of these features (diagenesis, overburden, geotectonic environment, heterogeneities, fluid-rock interactions, over-pressure)?

• How can we distinguish between permanent (steady), time-limited and short-term episodic flow in these features?

• What are the predominant factors for self-healing of preferential flow paths (rock strength, geotectonic environment, clay mineralogy, fluid-rock interaction)?

• What kind of investigations are recommended to answer the above mentioned questions?

• What can be learnt by experimental work? Do we get meaningful quantitative data? Do we increase process understanding?

• What can the radioactive waste, hazardous waste and petroleum industries learn from each other concerning flow through fractures and faults in argillaceous media? Which findings are transferable (analogous)?
Diagenesis and Compaction in Argillaceous Formations

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Extended Abstract

Argillaceous sediments are deposited in various nonmarine and marine environments. Their compositions, although variable, are dominated generally by >60% clay minerals with minor admixtures of quartz, feldspar, carbonates, organic matter and pyrite or iron oxides (SHAW & WEAVER 1965 [14]). Diagenesis of argillaceous sediments is dependent upon many factors, including depositional environment, types of clay minerals, amount of organic matter, pore water chemistry, geothermal gradient and burial history. The systematic loss of porosity recorded with increasing burial is related to gravitational compaction and chemical diagenesis, most importantly the progressive reaction from smectite to illite.

Following initial rapid porosity loss in the first 500 m of burial, porosity decreases more slowly, reaching 20-30% at a depth of 1000 m. As a result, the “open” depositional texture is transformed to a denser fabric of parallel oriented flakes associated with the expulsion of large volumes of porewaters (RIEKE & CHILINGARIAN 1974 [12], WEAVER 1989 [19]).

The physical reorganisation of clay flakes is followed during deeper burial by chemical compaction, including cementation, pressure dissolution and the conversion of smectite to illite. However, chemical processes begin during the early stages of argillaceous mud diagenesis. They are controlled by the depositional environment i.e. the composition and Eh of the interstitial waters. Whereas in the marine environment early pyrite is precipitated, siderite is formed in non-marine muds because of low sulfide activity.

Of the many diagenetic reactions which occur during deep burial of shales the most universal modification is the change in clay mineral composition through illitisation of smectite, transformation of kaolinite to dickite, illitisation of kaolinite and formation of chlorite. The smectite-illite transformation has been investigated by numerous authors since WEAVER (1959 [18]) first concluded that smectite is altered below 3300 m to mixed layer smectite-illite with the illite component increasing with further burial. Assuming potassium feldspar as a potassium source, the smectite-to-illite transformation can be described by the following reaction (ABERCROMBIE et al. 1994 [1]):

\[ \text{KAlSi}_3\text{O}_8 + 2\text{K}_0\text{.3Al}_1.9\text{Si}_4\text{O}_{10}(\text{OH})_2 \rightarrow 2\text{K}_0.8\text{Al}_1.9(\text{Al}_0.5\text{Si}_3.5)_0\text{Si}_{10}(\text{OH})_2 + 4\text{SiO}_2(aq) \]

K-spar  K-Smectite  Illite

This mineralogical change from swelling to nonswelling clays is associated with uptake of potassium, a two step dehydration and release of various cations such as Na⁺, Ca²⁺, Mg²⁺, Fe⁺ in
addition to silica. The cations released depend on the composition of the reacting smectite. The reaction is temperature controlled and, therefore, composition-temperature relationships depend on the geothermal gradient (VELDE & VASSEUR 1992 [17]; HILLIER et al. 1995 [10]) and residence time at temperature (EBERL & HOWER 1976 [6]; RAMSEYER & BOLES 1986 [11]). The starting composition of I/Sm mixed layer minerals is equally important. HARTMANN et al. [9] found that the I/Sm mixed layer minerals with a high initial smectite component (>60%) can be described by an S-shaped composition-temperature curve with an abrupt increase of the illite content at ca. 70±10°C. This increase is the result of an increased reaction rate and changes in ordering from R=O to R>1 (EBERL 1993 [7]) providing larger volumes of silica.

Although chlorite increases in weight percent with depth, indicating neoformation, it is generally more abundant in sandstones than in associated shales. While direct precipitation from pore fluids has been reported (BURTON et al. 1987 [5]), increase of grain size with depth and honeycomb arrangement of pore-lining chlorites in sandstones suggest chlorite formation via smectite-corrensite-chlorite (HILLIER et al. 1996 [10]), i.e. a sequence of precursor minerals. The observation of chlorite packets intergrown with smectite further supports such a mechanism (AHN & PEA COR 1985 [2]). Chlorite may also form, however, from kaolinite (see below) or biotite. Chlorite formation requires, in addition to other cations, ferrous iron, the most likely sources of which are dispersed hematite and other iron oxides. The reducing agents may be H₂S, CH₄ and liquid hydrocarbons, indicating the interdependence of inorganic and organic matter diagenesis (SURDAM et al. 1989 [16]).

Whereas kaolinite is frequently observed as an early diagenetic phase in sandstones, there is little evidence that it crystallizes in shales during burial (WEAVER 1989 [19]). Thus, when present, it is detrital in origin. At temperatures above ca. 70°C, kaolinite is converted to dickite. When exposed to porewaters with high K⁺/H⁺ or Mg²⁺/H⁺ activities (i.e. presence of K-feldspar), kaolinite converts to illite above 100-140°C and chlorite, respectively. The illitisation of kaolinite releases silica and water. Also in this temperature range, pressure solution and stylolitisation become important in sandstones and even more so in siltstones and shales, releasing large amounts of silica in particular.

Several authors, especially SCHMIDT & MCDONALD (1979 [13]) and SURDAM et al. (1984 [15]), suggest that carbon dioxide and carboxylic acids generated by bacterial and thermal decomposition of organic matter in shales greatly influence inorganic diagenesis. Water released during illitisation of smectite carries dissolved inorganic and organic ions into adjacent porous rocks, initiating precipitation-dissolution reactions and flushing out hydrocarbons.

Although there is ample evidence for the import of solutes from shales into adjacent sandstones, e.g. the presence of cementation fronts of quartz and calcite (FUCHTBAUER 1974 [8]), the quantity of solute expelled per unit volume of shale is not certain. BJ_RLYKKE (1983 [3]) convincingly showed that the porewater flux generated from the underlying sequence would be far insufficient to account for the amount of quartz cement normally observed in reservoir sandstones given low silica mobility. High resolution microscopy of shales indicates that many of the released ions may reprecipitate as cement within the argillaceous unit itself. WINTSCH & KVÆLE (1994 [20]) conclude from mass-balance calculations that most major oxides are immobile except K₂O and CaO which are added to or removed from shales, respectively, reflecting illitisation and dissolution of carbonate. Thus, shales may act as a source or sink for silica. According to BLOCH & HUTCHEON (1992 [4]) illitic shales become sinks of silica, not sources as diagenesis proceeds.
After late burial, shales invariably have porosities of less than 10% with permeabilities as low as $10^{-5}$ D, and ultimately show distinct fissility. Progressive porosity-depth trends may be modified, however, by overpressuring related to expulsion of fluids from compacting thick argillaceous formations, to dehydration reactions or to kerogen maturation.

Because diagenetic processes are linked to flowing fluids, which transport heat and ions, flow rates, duration of flow and fluid pathways are crucial. During shallow burial, porewaters are expelled from compacting argillaceous muds through the interparticle pore network. With increasing overburden and degree of particle orientation, accompanied by loss of porosity and permeability, flow is channelled along inhomogeneities such as sand layers, fractures and faults. Fluid inclusions often reveal precipitation of diagenetic minerals into fractures at temperatures less than 50°C, as well as the presence of injected clay fillings. Both indicate that fractures may form during shallow burial. Fractures are the single most important conduits in tight shales and largely control cross-formational flow.

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Faulting and Jointing in Sedimentary Rocks: 
Basic Concepts and Impact on Bulk Properties

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Abstract

In this introductory review, the more basic aspects of joint and fault analysis in little disturbed sedimentary sequences are discussed, with reference to recent literature and, particularly, to relevant experience from structural reservoir modelling in the oil industry. Analytically, jointing is relatively simple, since joints are, by definition, tectonic fractures (non-cohesive partings) on which no displacement has taken place. Joint populations are often systematic and can be treated statistically using simple orientation and size (usually trace length) parameters, taking into account problems of sampling bias. In contrast, faults - by definition, fractures on which significant slip has taken place - are more localized and irregular in distribution, more variable in orientation, and more complicated to parameterize because of the practical difficulties of defining the slip vector (and hence, amount of displacement). To a first approximation, small faults and microfaults can be thought of as forming an interconnected network in zones adjacent to large faults (the damage zone, which may or may not be accompanied by drag), and in other areas of strain concentration due to displacement variations and/or slip incompatibilities. These zones surround rock volumes of low fracture intensity. The scaling rules for the high-strain zones (damage zones including main faults) and the low-strain volumes (within the fault blocks) are different and this together with the irregular distribution patterns makes modelling difficult. However, structural modelling techniques have advanced considerably in recent years and now form the basis of sophisticated analyses of the influence of faulting on bulk rock properties for rock mechanical and hydrogeological studies, taking into account the physical changes which are typically associated with faulting. Much of the experience gained from the geomechanical and geohydraulic modelling of hydrocarbon reservoirs will be relevant to radioactive waste disposal in argillaceous rocks, keeping in mind the long time scale and the special characteristics of clay-rich material.

INTRODUCTION

This brief review covers some of the main ideas and problems related to faulting and jointing, particularly with respect to their geometry, distribution and impact on bulk rock properties in little disturbed sedimentary sequences. Little disturbed sedimentary rocks, as observed in surface exposures and near-surface excavations, usually show networks of planar discontinuities which are obviously potential planes of mechanical weakness and/or potential pathways for, or barriers to, fluid migration (cf. Price 1974, Hodgeson 1961, Hoshirio 1967, Lorentz et al. 1986 and many others). On a larger scale, they are often affected by faulting to some degree, in spite of the stable environment.
The aim of studying structural elements in waste disposal sites in such areas is to quantify and predict the fluid flow and geomechanical behaviour of the repository on both short and long term, based on geological concepts and using modern modelling techniques. To achieve this the terminology of features of interest has to be clarified and a parametrization has to be established to allow stochastic simulations and modelling approaches for testing. Usually, a grid model has to be established for later analysis which is based on structural elements and conditioned by their properties (Fig. 1). Large-scale elements detectable in the field and on seismic surveys define the boundary conditions for the host rock volumes. Smaller-scale features based on outcrop, core and well information are site specific, but are never known for the whole volume of interest. Therefore, careful mapping and characterization of these features is necessary to develop distribution rules and scaling properties, and to establish 3D models. Because it is impossible to fully test a repository site with regard to its fluid flow properties and mechanical behaviour, only proper modelling based on detailed analysis of structural elements will help to understand the influence of tectonics.

In the present paper, we summarize some of the more basic concepts related to fracturing in poorly consolidated sedimentary rocks ("soft" rocks, such as shale, poorly consolidated sandstone, and chalk) in layered sequences. Many of these concepts are derived from, or can equally be applied to, "hard" rock studies (limestone, granite, gneiss, etc.), but for space reasons we have not included the extensive literature on this aspect. Instead, we have attempted to encompass some of the literature from other fields of research concerned with "soft" rock tectonics and its influence on fluid flow and geomechanics, particularly as applied to hydrocarbon reservoir engineering, for instance, in the North Sea.

Fig. 1 Quantification of structural information and establishment of a grid model using on-site data and analogue data sets (after Koestler et al. 1995).
CLASSIFICATION

Rock discontinuities in sedimentary rocks fall into two main groups: primary – formed during deposition, before lithification (bedding planes, bedding fissility, formation boundaries, unconformities, etc.); and secondary - formed after deposition, during or after lithification. Here we focus on secondary discontinuities, which take the form of fractures related to compaction, exhumation and deformation, caused by differential movements in the Earth's crust and gravitational forces. These may be called tectonic fractures, to distinguish them from other fracture types which may be of non-tectonic origin (cooling joints in lavas, artificial fractures due to blasting, etc.) and which will not be considered further here. Tectonic fractures can be roughly subdivided into three main categories - joints, faults and veins/stylolites – depending on the observed movement of the fracture walls (cf. Pollard & Aydin 1988, Koestler & Milnes 1992, Dunne & Hancock 1994). Joints are fractures on which there has been no significant movement of the fracture walls, either laterally or perpendicularly, at the relevant scale of observation (for practical problems, usually with the naked eye in outcrop). There are some problems in applying this definition, as mentioned below, but it has in fact stood the test of time. Faults are fractures along which the fracture walls have undergone appreciable lateral displacement, at the relevant scale of observation, as evidenced by displaced markers and/or surface structures typical of slip (see below). Veins and stylolites indicate appreciable movement perpendicular to the fracture, veins in the opening and stylolites in the closing mode. The term vein implies mineral or magmatic infilling (an open fracture would be called a fissure) and is generally confined to small-scale features (large-scale magma-filled fractures are rather arbitrarily referred to as dykes and sills). Stylolites are formed by removal of material from the fracture site by pressure solution, and show characteristic features (Ramsay & Huber 1987) – if these features are absent, they are referred to as pressure solution seams. In this review, we concentrate on joints and faults, as the structures most likely to be mechanically and/or hydraulically significant in the context of clastic sedimentary rocks. However, the local importance of, for instance, partially filled veins or clay-rich pressure-solution seams, should be kept in mind.

In the following, we will initially take a phenomenological approach, describing first the main physical features of single joints and faults, as they are observed in the field. This is important from a practical point of view, because many of the uncertainties in joint and fault analysis are intrinsic to the phenomena and not due only to sampling difficulties. The main parameters required for quantitative analyses and structural modelling will then be discussed, followed by an overview of the types of statistical analysis which are most often employed. These sections will be subdivided into a discussion of orientation parameters and analyses, on the one hand, and size parameters and scaling, on the other, and the problems associated with each, including comments on hydrogeological and/or geomechanical significance. Finally, we consider some aspects of the modelling of joint and fault systems in little disturbed or extended sedimentary sequences.

TYPICAL FEATURES

Both joints and faults can be idealized to a first approximation as planar features of finite extent. Finite extent means that fracture planes are surrounded by a fracture front, or tip line, outside which the rock is unfractured. This idealization is used as a basis for defining orientation and size parameters (see below), but in fact is seldom observed in Nature. Almost all fractures show some degree of roughness (an important element in stability evaluations), and also curves and kinks on a larger scale, and many show characteristic markings on the fracture surface. On the other hand, very few fractures show well-defined tip lines, even where isolated (i.e. not interfering with neighbouring fractures), but rather show branching patterns, or fringes (Fig. 2).
Joints and jointing

Joint populations of presumed tectonic origin are ubiquitous in apparently undeformed, sub-horizontal, sedimentary sequences (e.g. Price & Cosgrove 1987, Cruith Shank & Aydin 1995), but their genesis is problematic and controversial. They are generally sub-vertical, often bedding-confined, and usually arranged in joint domains of different or systematically changing preferred orientation, showing little relation to large-scale structures (major faults or folds). Well-developed joint sets often show different characteristics in different lithological units, apparently related to the different mechanical properties of the rock types (mechanical stratigraphy e.g. Corbett et al. 1987, Gross & Engelder 1995) and to variations in bed thickness (thickness-spacing relationship e.g. Mandal et al. 1994, Wu & Pollard 1995).

Joint surfaces show typical associations of plumose markings, conchoidal rib marks and fringes of en echelon minor joints (Fig. 2), which can be interpreted in terms of the established principles of fracture mechanics (Pollard & Aydin 1988). The point of convergence of the plume striations, or hackle marks, represents the origin, or point of initiation of the joint, the axis of the plume marks the leading tip of the fracture front, and the opening of the plume points in the direction of fracture propagation. These features are typical of mode I fractures, i.e. extensional fractures, with (very small) opening displacements perpendicular to the fracture surface, but there is disagreement as to whether similar markings would be produced in the shearing modes (fracture modes II and III) or not.

Fig. 2 Fringes at the edge of a joint surface (Entrada Sandstone, Utah, USA).

Faults and faulting

In contrast to joints, faults are more localized and irregular, both in orientation and distribution, and the faulting process physically changes the rock properties along, and in the vicinity of, the fault (Fig. 3). Although an artificial subdivision, it is convenient to distinguish different scales of structure: regional faults (major structures, with displacements measured in kilometres and upwards), large faults
(displacement of 10s to 100s of metres, visible on seismic reflection profiles), **small faults** (displacement of centimetres to metres, visible in outcrop but not on seismic sections) and **microfaults** (displacement of millimetres, visible in hand-specimen, core or thin-section). Regional and large faults are characterized by zones of fracturing, brecciation and gouge formation (Sibson 1977, Scholz 1987, Laubach 1988) and, in shale-bearing sequences, clay smearing (Weber 1978, Gibson 1994, Koestler & Fjaersto in press.), which together with effects due to the juxtaposition of different lithologies across the fault (Smith 1966, Nybakken 1991), strongly affect fluid flow paths, either as barriers or as conductive zones. At the other end of the spectrum, microfaults show fabric changes, e.g. in sandstone, grain rotation and grain-size reduction, forming “deformation bands” (cf. Aydin & Johnson 1983, Gabrielsen & Koestler 1987, Antonellini & Aydin 1995), in shale, homogenization and porosity reduction (e.g. Berg & Avery 1995), and sometimes mineralogical changes, which, when compounded in large populations, significantly change the bulk rock properties. In faulted sedimentary sequences, poroperm relations and rock mechanical variations show distributions which are significantly affected by the geometry and kinematics of the large fault network and the arrangement of small faults and microfault populations in the intervening blocks. Since the tensile strength of rocks is significantly lower than the compressive strength (cf. Lockner 1995), fracturing in little disturbed sedimentary rocks is often determined by the action of small, horizontal extensional strains, both on a large scale (normal faulting) and on a small scale (extensional jointing and microfaulting).

**Fig. 3** Small-scale fault in Jurassic loosely-consolidated sands (Bornholm). Due to displacement of about 1 m, the rock fabric within the fault zone is changed so that the zone is sealing for coloured groundwater (dark brown staining to the right hand side, footwall). At microscopic scale, denser physical packing of sand grains can be observed. Clay smear along the fault also contributes to increased sealing capacity. (width of photo ca. 1m)

Fault surfaces, on which appreciable slip has taken place, show markings and physical features related to the movement of the fracture walls, over printing features formed during the phase of fracture initiation and propagation. The most obvious marks are generally slickensides or slickenlines (Ramsay & Huber 1987, Hancock & Barker 1987), which, because they are known from experiments to form after only small amounts of slip, may represent just the very last increment of the total movement on the fault. Slickensided surfaces may be covered with purely mechanical grooves or arrays of aligned, fibrous crystals, associated with steps and chatter marks, or an overlapping arrangement of fibres, and these are often used for determining the sense of movement, in the absence of displaced markers. Other features include pockets of crushed material (fault gouge, fault breccia), drag and/or smearing of wall rock along the fault, and other features, which, on a large scale, become important elements in a major fault system. The fringes around faults are more complex and variable than joint fringes, often associated with en
echelon systems of tension gashes, where the concentrated shear displacement on the fault plane is translated into more distributed shear in the tip region, or with asymmetrical zones of extension and shortening as the displacement decreases.

![Diagram](image)

**Fig. 4** As an example, an extensional fault block is subdivided into high-strain zones (fault and damage zones) and low-strain volumes.

To a first approximation, small faults and microfaults can be thought of as forming an interconnected network in a zone adjacent to a large fault, the “damage zone” (Chester & Logan 1986, Koestler & Milnes 1992, McGrath & Davison 1995, see Fig. 4). Damage zones develop against even ideally planar faults in, for instance, massive sandstone and chalk, due to a combination of fault tip propagation and frictional resistance to movement on the fault plane. In well-bedded rocks and shale-dominated successions, ductile deformation and flexural-slip reduce the need for fracturing by producing a “drag” zone (Hamblin 1965, Scholz 1995, Reches & Eidelman 1995). The zonation of bulk rock properties associated with a single large fault can thus be described in terms of the fault zone itself (breccia, gouge, clay smear), the damage and/or drag zone (which extends beyond the fault tip lines), and the surrounding undeformed rock (Koestler et al. 1995). This simple zonation can also be expected in a system of interconnected non-planar faults which are slip-compatible (i.e. in which all faults contain the same slip vector cf. Koestler et al. 1992), in which case the fault zones and damage/drag zones would surround undeformed rock volumes in each fault block. However, most systems of large faults in layered sequences are not slip-compatible, due to lithological control (e.g. alternating competent and incompetent horizons) and vertical and horizontal variations in stress/strain regime. Fault bends and non-systematic displacement variations are the general rule (e.g. Peacock & Xing 1993, Huggins et al. 1995), and the fault blocks show extensive accommodation to incompatible movement and rotation (relay zones, fault-bend folds, “reverse drag”, roll-over anticlines, etc.). This appears as populations of small faults and microfaults, or ductile deformation (depending on lithology, pore pressure, stress regime, etc.), associated with irregular variations in bed orientation. Much progress has been made in recent years in systematizing these complex patterns of fault-related fracturing and block-internal deformation, based on detailed study of field examples, and in incorporating this experience in modelling procedures for fluid flow studies (e.g. Koestler et al. 1995, Makurat et al. 1995).

**PARAMETRIZATION**

In addition to the observation and understanding of the physical nature of the structures, joint and fault analysis is based on the statistical treatment of individual measurements made in the field (natural outcrops, tunnels, quarries, drill cores, well bore images, etc.). Measurement and sampling problems make up a significant part of the uncertainty attached to such analyses. The main parameters used in joint and fault analysis can be subdivided into those defining orientation, those defining dimension (size) and those defining hydraulic and/or mechanical properties. These are treated separately below.
Orientation

The basic orientation parameter is the strike/dip of the fracture plane (or in 2D analyses, on horizontal or vertical surfaces, either the strike or the dip). In the case of joints, this is usually the only measurable parameter, unless well-developed plumose markings and a particular application make a detailed analysis of joint propagation directions appropriate. In contrast, faults require, in addition to the orientation of the fault plane, also the slip vector for complete definition. The slip vector (usually defined as the movement direction of the block overlying the fault - the hanging-wall - relative to the underlying block - the footwall) is a representation of the net slip on the fault, deduced from the observed displacement of markers and/or from the surface markings (slickensides, etc.), assuming a constant movement picture. The slip vector is a combination of a linear parameter (slip direction, with a measured plunge/trend) and a polarity or “sense of shear” parameter. In the case of slickensided fault surfaces, it is the polarity of movement which is difficult to determine. In other cases, for instance, interpreting seismic data, the problem is to determine the slip direction. Hence, the slip vector is often difficult to determine precisely, but under favourable exposure conditions it becomes the key element in fault slip analysis (Angelier 1989, 1994, Petit & Mattauer 1995), i.e. for determining the paleostress field (using the same inversion techniques as applied to earthquake focal mechanisms).

Dimension (size)

Parameters defining dimension (or size) are even more problematic. For both joints and faults, many workers use a “length” parameter, often the trace length on a 2D section (aerial photograph, structure map, quarry face, tunnel floor, etc.). The measurement difficulties here are formidable. The trace length depends on the position of the section relative to the fracture (the “Schnitteffekt”), and the fringes often appear as numerous, separate, small fractures. Also, many fractures do not have a tip line but intersect or merge with neighbouring fractures. In many cases, one or both ends of the fracture trace lies outside the observation window and the measurements have to be coded accordingly (censoring, e.g. Watterson et al. 1996). In some common situations (drill cores, sometimes in tunnels), fracture tips are only rarely observed and a length parameter makes no sense. An additional problem in quantifying the length of a structural element is the fact that size recognition is related to scale of observation. Tectonic structures continue usually along their strike direction below the scale of observation whether we are studying a satellite image, an outcrop or a thin-section. To scope with this problem modern mathematical techniques have been applied to describe scaling properties of different features (e.g. Mandelbrot 1967, Feder 1988, Koestler et al. 1995, Keller & Koestler, in prep.). All in all, size statistics based on length are subject to a great deal of uncertainty.

For faults, however, a more reliable size parameter is available - maximum displacement, or net slip in the central region of the fault plane (Walsh & Watterson 1988, Cowie & Scholz 1992, Dawers & Anders 1995). On land, this parameter is no less intractable than length, although, under some circumstances, it may be a determinate quantity. However, in a good quality 3D seismic survey, some component of maximum displacement (e.g. throw, separation) can often be determined for many faults in one area, and, assuming a constant slip vector, can be used for statistical analyses. Another size parameter which has been suggested for faults is the thickness of the associated zone of fault gouge and breccia (e.g. Knott 1994 and many others), related, of course, to the lithology of the wall rock and other factors.
Hydraulic and mechanical properties

To use structural information in flow models, understanding of the hydraulic and mechanical properties of the individual structures involved is necessary. In the same way as the bulk properties of the host rock has to be established from well logs, core samples and analogue sources, the properties of deformation elements need to be quantified. The testing of individual structures is often difficult because of their wide heterogeneity, the difficulties of direct measurement and sampling problems. Mechanical apertures of joints are critical for fluid flow in jointed rock masses, but they have to be related to joint stiffness and joint roughness in terms of their stress dependency (Barton et al. 1985, Marsily 1985, Braun & Scholz 1986, Lorenz et al. 1986, Makurat et al. 1995). This is also true for fault rocks, such as breccia and gouge (cf. Morrow et al. 1984): the capillary pressures of the deformation products can give a measure for their flow properties which can be used in advanced simulators. Faults consisting of fractures of different types have to be characterized not only in their specific and individual properties but also in their network properties including geometry, density, interconnectivity and variability. This will be returned to below after a consideration of the different types of statistical analysis.

STATISTICAL ANALYSIS

The collection of individual measurements of (preferably individually described) fractures in sufficient numbers leads to semi-statistical analysis, to outline the broad patterns and general features of the fracture system and to assist in the construction of a structural model. The term "semi-statistical" indicates that geology only rarely lends itself to true statistics, due to the non-systematic nature of outcrops and the time-consuming nature of data collection (Fig. 5). These generally result in inadequate data sets for which specific mathematical tools have been developed (e.g. Swan & Sandilands 1995). Joint and fault analysis usually addresses inverse problems: the construction of models based on the analysis of small (inadequate) samples, combined with general structural geological knowledge and experience (e.g. Yin & Ranalli 1995). In this connection, different types of diagram are common in the literature and serve as a basis for the following discussion.

Spatial distribution

Orientation data are plotted as a histogram or rose diagram, or as a stereogram, depending on whether the data are 2D or 3D in character. A rose diagram is simply an histogram plotted in polar coordinates and has certain visual advantages (e.g. geographic orientation of the main peaks) and some disadvantages (e.g. exaggerates the importance of the main peaks). Both the histogram and the rose diagram show the frequency of fractures occurring within different orientation intervals (usually 10°). Some confusion arises in the literature because two frequency parameters are in general use and are not always distinguished. One shows the number or percentage of measurements per angular interval and is usually used for the representation of unsystematically collected field data, or for scanline or drill core data. With sufficient data points, it brings out the general outlines of the pattern (e.g. number and orientation of joint sets), but the relative height of the peaks has no significance. The other type of data representation shows the cumulative length of fracture trace per angular interval, and results from the systematic measurement of fracture trace maps in areas of complete exposure (for instance, digitized satellite images, fault maps and outcrop grid nets). In this case, the relative lengths of the columns have a statistical significance, and the data are amenable to other types of treatment (e.g. a fracture density parameter can be defined for each set as the total length of fracture in that set per unit area). The techniques for processing orientation data from linear profiles (scanlines, cores, etc.) and areal surfaces
(fracture trace maps) are discussed in detail in La Pointe & Hudson (1985, see also, for instance, Annels & Hellewell 1987, Milnes & Gee 1992).

Fig. 5 Regional joint sets with different orientations defining blocks of little deformed or undeformed rock volumes. Part of San Raffael Swell in Utah.

The stereogram is the traditional way of plotting 3D structural data. With a large amount of data, planes are generally plotted as poles and presented as pole diagrams, or their contoured equivalents, or (preferably) both, using various statistical techniques, today mainly computerized. 3D data are generally unsystematically collected, but are assumed to illustrate the main trends, when these are clearly visible. The computer programmes usually determine, for instance, the mean pole for an individual pole maximum distribution (clusters of poles), or the mean great circle for a girdle distribution. In many cases, however, the results of using such statistical methods needs to be scrutinized critically to avoid over-interpretation. Critical use of stereographic projection is particularly important in fault slip analysis, where the usually small amount of data consists of coupled orientation parameters (planes containing lines of defined polarity, Angelier 1989, 1994). “Black box” statistics in such cases may be quite inappropriate (Huang 1988).

Scaling rules

Statistical treatment of the different size or dimension parameters play an important role in defining the rules and distribution functions in the modelling of joint and fault systems. For review purposes, these can be subdivided into three groups. The first group consists of analyses relating the different size parameters to each other. The most common are discussions of the relationship between fault length (for instance, in map view) and maximum displacement (e.g. Walsh & Watterson 1988, Cartwright et al. 1995, Dawers & Hancock 1995), and between length and/or displacement and the thickness of the fault zone (e.g. Hull 1988, Knott et al. 1996). These analyses are important for the integration of different types of data (fault maps, seismic sections, core/borehole data, etc.). A second group deals with the statistics of fracture spacing, as one of the main parameters for hydrogeological or geomechanical studies (ISRM 1978, Priest & Hudson 1981, Hudson & Priest 1983, Narr & Lerche 1984, Sen & Kazi 1986, Aguilera 1988). In sedimentary rock sequences, some attention has been paid to the relation between spacing and lithological variations (“mechanical stratigraphy”, cf. Corbett et al. 1987) and/or bed thickness (e.g. Wu & Pollard 1995, Fischer et al. 1995). A third group of

Fig. 6 Log-log plots of cumulative number of faults vs. Length indicating changes in scaling rules over the scale range of seven orders of magnitude. $10^2$ - $10^7$. (see text for explanation)

Spacing and length distributions follow rules of rock fracture behaviour as a result of mechanical properties and applied stresses. Size distributions of faults or magnitude frequency of earthquakes, are generally accepted to behave in a scale invariant manner (e.g. Gutenberg & Richter 1942, Koestler et al. 1995, Walsh & Watterson 1996, and many others). These empirical observations are used to establish scaling laws for the prediction of information at scales which are not directly observable (e.g. Scholz & Cowie 1990, Walsh et al. 1991, Gauthier & Lake 1993, Needham et al. 1996, Watterson et al. 1996). We have collected data from intraplate extensional areas and plate boundaries, and combined them with published data, such that fault length is continuously represented over seven orders of magnitude (Fig. 6). After corrections for different types of sampling biases, scaling behaviour was found to be constant within three distinct length ranges. Whereas large and small fault lengths obey the same scaling law (as represented by a straight line in log length vs. log cumulative number plots), the medium length range (~50m-500m, equivalent displacement ~0.5m-20m) scales distinctly different, indicated by a lower fault frequency compared to other lengths. This leads to a gap in sample sizes (i.e. a smaller number of faults within the scale gap than predicted following the power law) of about one order of magnitude. The existence of a scale gap was first observed and documented from field areas (Koestler et al. 1995) and seems to find its explanation in stress impact on layered heterogeneous rock sequences.

MODELLING OF JOINT AND FAULT SYSTEMS

The aim of the description, parametrization and statistical analysis of structural phenomena, and the integration of different types of structural, mechanical and hydraulic data, is to aid in the development of relevant models for simulating physical rock/fluid interactions in the subsurface according to the problem at hand. With regard to sedimentary sequences, much of the experience in this field comes from
hydrocarbon reservoir geology and engineering (e.g. Brand & Haldorsen 1988##, Koestler et al. 1994, and many others) and will need to be modified to encompass the long-term effects relevant to radioactive waste disposal. For all applications, however, models can be thought of as constructed out of three main components - structural model, geomechnical model, geohydraulic model - which together aid in estimating bulk rock properties and their variation in space and time.

**Structural model**

The structural model is a representation of the 3D distribution of the structural elements within the rock volume of interest, together with an elucidation of the structural history (sequence of events). As indicated earlier, brittle failure of sedimentary rocks under crustal extension can often be modelled as consisting of two structural environments, major normal fault zones along block boundaries, and distributed small-scale fracturing within the intervening blocks (e.g. Koestler et al. 1994, Koestler et al. 1995) (Fig. 7). The fault zone model includes descriptions defining variations in the width of the fault zone itself (gouge, breccia) and the related damage zone, both belonging to the high-strain zone, which are related to displacement on the fault. In addition, the influences of fault bends, intersections and relays on overall fracture distribution have to be estimated and quantified (e.g. Peacock & Xing 1993, Gibson et al. 1994, Koestler et al. 1995), as well as the history of faulting (e.g. Davison 1995). The block-internal deformation will be represented by fracture systems modelled with input from in-situ and analogue field data (e.g. Chiles 1988, Oda & Maesibo 1985, Koestler et al. 1994, Koestler & Reksten 1995). In some tectonic situations, this two-part subdivision in high-strain and low-strain rock volumes may not be relevant, but the general philosophy of structural modelling will be the same (see Milnes & Koestler 1994).

![Fig. 7 Simulated damage zones to a structural larger-scale element. Simulation based on analysis of well data and established scaling rules.](image)

**Geomechanical model**

The geomechanical model outlines the dynamic significance of the structural model in terms of past, present and future stress and strain. An understanding of paleostress/paleostrain conditions at the time of generation and reactivation of the fracture systems (e.g. Angelier 1994, Krantz 1988, Petit & Mattauer 1995) is often critical for the refinement of the structural model or its extrapolation to areas of no or poor data. A knowledge of the present in situ stress field and its probable effect on already formed fractures in different orientations is necessary for understanding the geohydraulic significance of the fracture system (e.g. Barton et al. 1985, Fischer et al. 1995, Lorenz et al. 1986). And for the geomechanical modelling of the underground excavations. Geomechanical considerations are also critical for predicting the stability of bedrock sites under expected future stress fields (e.g. Stephanson & Shen 1991, Israelsson et al. 1992, Nieto-Samaniego & Alaniz-Alvarez 1995).
Geohydraulic modelling

For most applications, including radioactive waste disposal, the aim is to predict the geohydraulic significance of structural and geomechanical models for subsurface fluid flow, based on the observed hydrogeology of the site and on the determined or assumed hydraulic properties of the structural features. Detailed considerations of these models lies outside the scope of this review, and, for argillaceous rocks, will form a main focus of the present workshop. However, key themes are likely to include evidence of past hydrothermal circulation systems; identification of present-day water-conducting features; in situ or in laboratory measurements of the permeability of fracture types, fractured rock and fault rocks; consideration of the geometrical effects of fault displacement (juxtaposition, membrane effects); and other aspects determined by site-specific features (Antonellini & Aydin 1995, Jamison & Stearns 1982, Berg & Avery 1995, Gibson 1994, Zhang & Sanderson 1996). These themes precede scaling, ranking and homogenization procedures as a prelude to simulating system evolution using variations in bulk rock properties (Koestler et al. 1995).

Fig. 8 Simulation of the fracture geometry within high-strain zones and low-strain volumes and their influence on the fluid-flow in host rock compartments (presented as 100 - residual permeability to show the barrier effect of structures), see Koestler et al. 1995.

CONCLUDING REMARKS

This short discussion has attempted to filter out those aspects of structural geology and applied tectonics which are of particular relevance to an understanding of fracturing in relatively undisturbed sedimentary sequences and its geomechanical and geohydraulic significance. Much of the experience in this area comes from the oil industry, but, wherever possible, the paper has been tailored to the workshop theme - radioactive waste disposal in argillaceous formations. In this context some special characteristics of clay-rich rocks (clay, mudrock, mudstone, shale, marl) should be kept in mind. Firstly, compaction is an extremely important part of the evolution of clay-rich formations, causing the successive reduction in porosity (and rock volume) from perhaps 70% in waterlogged sediments to less than 10% in some indurated shales and marls. At the same time, argillaceous rocks attain a fissility (preferred orientation of clay minerals) and often also compaction-related fractures which has a significant effect on the rock's geomechanical and hydrogeological properties. The degree of pore-closure and dewatering, and the depth of burial, determine the mechanical properties of burial of clay-rich rocks in a complicated way which leads in some situations to ductile/plastic deformation (décollement, diapirism, smearing along faults, e.g. Hasse et al. 1985) and in others to brittle deformation (hydrofracture, jointing in unconsolidated clay, Price & Cosgrove 1987). Clay smearing along discontinuities significantly changes hydraulic properties of rock zones for both cross-flow and along-flow, mainly in a reducing manner (e.g. Koestler & Fjaerstoft 1996, Knipe et al. 1996) but also in flow increasing way especially when clay reacts brittly during reactivation and uplift of former deeply buried ductile sediments.
These, however, are special effects which will be a main focus of the workshop. In this introductory paper we have taken a wider view and attempted to summarize some of the more basic aspects of joint and fault analysis in little disturbed sedimentary sequences.

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Relevance of Faults and Fractures to Fluid Flow in Argillaceous Rocks

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Abstract

This paper attempts to summarize for researchers with widely divergent backgrounds some of the general aspects and problems of fluid flow along faults and fractures in argillaceous rocks for repository evaluation. The general topic is addressed by considering:

- Present knowledge of the generic condition of fabrics and geometries of faults;
- implications of fault fabrics for fluid flow and flow anisotropy;
- unique physical and mechanical properties of shales and other argillaceous rocks;
- modifications of fault fabrics and geometries as they cut through argillaceous rocks, and the subsequent influence on their potential as fluid conduits and/or seals;
- and observations of chemical interactions in shales along faults as a result of fluid flow.

Although the characteristics of single faults have been studied, they seldom occur as isolated features but generally as part of a system of common origin. Fault-zone geometry is complex, but in section parallel to the displacement, generally a fault rock of comminuted material is encompassed in a damage zone of fractured dilatant rock. A predictable fabric develops in the fault rock composed of fractures. Fault slip is localized along these fractures. Flow parallel to the fault is facilitated in the damage zone and fractures within the fault rock, but flow at high angles to the fault is inhibited by the fine grain-size of the fault rock. Thus faults may act as both conduits or barriers to fluid flow on the same fault varying in time and space. Cycles of fluid flow along faults reflect these changes with time.

Argillaceous rocks vary widely in their phyllosilicate content. Many of the shales in the world contain 90-95% quartz and have markedly different physical and mechanical properties from those that contain 45-65% quartz and 40-45% phyllosilicates. The latter are often perceived to be weak and ductile by comparison to most other rocks, but when tested in the laboratory in a nominally dry condition show little differences in strength and ductility from most sandstones and limestones. It is only when saturated that their strength drops to 6-25% of the dry state, but surprisingly they have not been found to undergo significantly further decreases with increasing temperatures to 225°C, nor does their ductility appear to be significantly enhanced.

Fault-zone fabrics in clay-rich materials show similar geometries to those in other rock types, but the alignment of clay particles defines the fabric, thus most flow along the fault is accommodated by the damaged zone. Flow across the fault is severely inhibited. The potential for
such faults to act as seals to fluid migration either across, or along them is greater than in other rock types.

Chemical interactions between fluids and fault rocks offer potential modifications of the fluid transport properties. Mineral solution and deposition within faults have been observed to “heal” the fault, thus decreasing potential for slip and subsequent fluid flow. Often, however, the result is a more brittle material which upon failure develops fractures and faults of relatively high permeability. Fluid transport through the fault may remove material, widening fracture apertures and facilitate higher volumes of flow.

Areas of significant uncertainties remain and require substantial research such as: further investigations of under-compacted argillaceous rocks to relate composition to mechanical and physical properties and both single and two-phase flow; scaling of laboratory measurements to natural conditions; and the interaction of fault systems and fluid flow as opposed to flow along a single fault. Such a program is optimally done through an integration of laboratory studies, field measurements and modeling efforts.

**Introduction**

Issues of fluid flow in argillaceous rocks that are cut by fractures and faults pose particularly difficult questions. These center on the potential of these features to act as conduits or barriers to fluid migration and how the properties of shales, siltstones and similar rocks alter the phenomenon. In general, our knowledge of the fluid-transport properties of faults and fractures is empirical and fragmentary at best. Shales, claystones and siltstones, although accounting for over 75% of the sedimentary column, have poorly documented physical properties at conditions above surface temperature and pressure, and over the time frames of interest. This situation is exacerbated in that compositions, textures and fabrics of these rocks, which control the physical properties, are seldom obtained when the latter measurements are made, making comparisons between data reported by different workers difficult.

These problems become evident when one anticipates locating repositories for radioactive waste in such environments. In that the requirements for such facilities are complex, they require careful evaluation as to the current state of knowledge and potential research requirements. On the basis of that evaluation, future work can be planned and will, of necessity, require an integration of laboratory studies, field measurements and modeling efforts. A goal will be to acquire pertinent hydrologic information combined with physical and mechanical data on faults and the relevant rock types for times up to 1000 years. The use of such data to create models should aid in predicting the response of a repository to a range of anticipated conditions, assist in design of adequate means of monitoring the repository, and if necessary, plan accessibility to the site for many years.

Generally, the issues of concern are: under what circumstances do seals to fluid migration exist; what conditions would threaten their integrity; if breached can they heal; and if so, in what time frame. Here the term seal is used as: “an interval of relatively low permeability, restricting fluid flow over the time frame of interest--from engineering to geological intervals”. Typically, seals are either roughly horizontal or approximately vertical. The former are often coincident with lithologic units while the latter are often defined by faults. Seals can be characterized by:

- the sealing interval is generally thin with respect to the surrounding volumes of rock;
- the result of restricted permeability is manifested in different fluid pressures, fluid composition, etc. between rocks on either side of the seal;
- a seal may vary in thickness from millimeters to tens of meters;
- seals may extend up to tens of kilometers either horizontally or vertically;
it may be homogeneous in lithologic character--composition, texture, fabric--or be heterogeneous at all scales;
although seals have commonly been placed in argillaceous rocks they are not restricted to that composition but have been postulated for a range of lithologies;
a seal is seldom a zone of zero permeability, even over engineering time scales;
seals are rarely static in time and space with cycles of rupturing followed by healing having been documented.

This paper only attempts to highlight some of the areas and problems of interest and illustrate some of the data available; in no way does it attempt to survey all of the existing literature. It approaches the general topic of fluid flow along faults and fractures in argillaceous rocks by first discussing the general conditions and then modifications to the general case pertinent to the specific problems of this conference, and from the simple to more complex, by considering:

our present knowledge of the general geometries and fabrics of faults and their implications for fluid flow;
the unique physical and mechanical properties of shales as representative of some argillaceous rocks;
anticipated modifications of fault fabrics and geometries as they cut argillaceous rocks and their subsequent ability to enhance fluid migration or act as seals or barriers;
a few observations of chemical interactions in shales along faults resulting from fluid flow.

Finally, a few thoughts on future research are presented.

The Influence of Generic Fault Geometries and Fabrics on Fluid Flow

Although the geometry and fabric of individual faults have been studied with subsequent inferences for fluid flow, faults seldom exist as single entities but generally occur as components of a genetically related system. It is the system, therefore, that needs to be considered as individual faults may have had varying histories leading to differences in properties of the damage zones and fault rocks [1-4] It has been observed that while many faults in a given system show evidence of past fluid migration not all faults appear to focus fluid flow, some appear to have acted as seals over at least part of their area and during their history. The reasons for such disparity are not clear at present; this is an area where more research would be instructive.

The geometry of individual faults is complex in both strike and dip directions, however, studies have established a general cross-sectional geometry that has been found to typify many faults within the upper portion of the crust [4-6]. The nominal fault zone width, or that which is often mapped as the "fault" on the ground, is a damage zone where the fracture density is significantly higher than the country rock. Within this is the fault rock or gouge, a region of fine-grained material that is much narrower than the damage zone, often only 5-10% of that width (Figure 1) [7, 8]. The fault rock has been deformed by a variety of mechanisms depending upon the pressure and temperature regimes. These range from mechanical comminution, "smearing" of more ductile material, dislocation glide and creep, recrystallization, to solution transport. Increasingly, it is recognized that more than one mechanism operates in a given environment, with one dominating for an interval but being supplanted by another at times, frequently cycling back and forth [9].

It is within the gouge or fault rock that the displacement is accommodated; this is the "active trace". Laboratory experimental work on simulated gouge has established a characteristic fabric that has been verified in many natural faults (Figure 2) [3, 10-13].
Within the fault rock, the grain size is significantly smaller than in either the damage zone or country rock as cataclasis by microfracturing with rotation and translation of the grains is a primary mechanism within the upper crust. The grain-size reduction is not homogeneous across the gouge but is localized. This is documented in experimental studies on simulated gouge of quartz which show that the average grain size is reduced by an order of magnitude after an initial shear strain of about 20%. Along the early formed R₁, R₂ and P-fractures they are reduced by another factor of ten, and along the Y-fractures a third order of magnitude [14]. It is along the latter that displacement eventually becomes localized when the shear strain reaches some critical value. This produces a relatively steady-state friction-time relation [12, 15].

The result of the grain-size reduction is that the porosity and permeability are heterogeneous within both the fault rocks and the fault zone producing a condition of flow anisotropy [8]. Fracture porosity and resulting permeability are highest in the damage zone, higher than in the relatively unfractured country rock adjacent to it; fluid flow parallel to the fault is preferentially enhanced in this domain. Additionally, flow is enhanced by the presence of fractures within the fault rock, especially those of Y orientation. The fractures are sufficiently open so fluids can potentially migrate along them. The result is that parallel to the fault-slip direction fluid flow is not only enhanced within the damage zone but may also occur within the fault rock along these fractures. Transverse to the fault rock, however, restricted flow results from a decrease in grain size with a corresponding increase in the tortuosity of the migration path. Additionally, there is a lack of major fractures crossing the fault rock once Y-fractures are well developed. Thus the fault rock is potentially a barrier to fluid flow often forming a seal to fluid flow at high angles to the fault. The occurrence of faults that act as seals to fluid flow across them has become a common feature of many hydrocarbon basins and is now well documented by pressure and in some cases geochemical differences between fluid compartments.

This restricted flow may also produce significantly higher values of pore pressure within the fault rocks than even the adjacent country rock. The domains partitioned by barriers of low permeability along R-, P- and Y-fractures may produce a series of pressure compartments [12]. This can result in differences in pore pressure from the center to the periphery of the fault rock, or along strike, and can lead to pore pressures above hydrostatic. Such conditions have been encountered in drilling in hydrocarbon reservoirs and may produce effects similar to those proposed by Rice [16] and Byerlee [17] to alter the state of stress within the fault zone.

To indicate the potential magnitude of permeability reduction caused by even a small amount of shear strain, measurements on quartz gouge with a starting grain size of about 1 micron and sheared about 5 mm at an effective confining pressure of 150 MPa, resulted in permeabilities in the micrdarcy range as measured across the zone [18]. In another set of experiments, cylinders of Berea sandstone containing a saw cut parallel to the loading and fluid-flow directions were taken to failure in triaxial compression where a fault formed intersecting the saw cut. The fault zone was about one millimeter thick and underwent an equal amount of displacement. Air permeabilities decreased from about 850 md to about 250 md or about 30% or their original value with this very small deformation [19]. These experiments suggest that within fault rocks even where the displacements on R-, P- and Y-fractures are very small, significant decreases of permeability may occur.

The preceding observations can be summarized as:

- Faults are generally weaker than the surrounding rock.
- Faults seldom occur as isolated features, but most frequently as parts of genetically related systems, so both must be considered in evaluating fluid flow.
- Faults can act as conduits or seals to fluid flow; they can be both at the same time in different parts of a fault or fault system and they may change in time.
• Not all faults enhance fluid migration nor act as a seals, some appear to have little affect on fluid migration.
• Fault geometry is complex both in strike and dip sections.
• Damage zones and fault rocks generally have widely different properties.
• The fabric of fault rocks is seldom random and is often predictable which allows estimations of it's contribution to fluid flow.
• Faults may often be anisotropic to fluid migration; flow parallel to the fault trace is enhanced in the damage zone and along fractures within the fault rock, but inhibited at high angles to it.
• Fault properties are not homogeneous along a fault, nor are they static in time. The fabric may be altered by mechanical processes, chemical interaction of fluids and the rock or most likely some combination of the two.

**Mechanical Response of Argillaceous Rocks**

Argillaceous rocks vary widely in their phyllosilicate content, composition, and grain size, ranging from poorly consolidated claystones to siltstones to shales. Concurrent with these variations are differences in mechanical and fluid-transport properties. As shales are an abundant end member of these rock types, they have received considerable attention.

The largest volumes of the shales in the world occur in the lower Paleozoic and are composed of 85-90% quartz and consequently have markedly different physical and mechanical properties from younger rocks also classified as shales. The latter commonly contain about 45-65% quartz and feldspars, and 40-45% phyllosilicates. Because of the relatively high clay content they are commonly perceived as being weak and ductile by comparison to most other rocks and have often been invoked to form seals to fluid flow either along faults or as stratigraphic units. Conversely, field observations show some to have sustained significant fracturing with fluid migration and mineralization. Natural hydrofractures in shales produced by pore pressures approaching lithostatic have been credited with producing “leaky seals”.

Laboratory studies demonstrate the extremes in mechanical behavior. Probably the most significant parameter is the ratio of clay species to quartz plus feldspar. With increasing amounts of quartz and feldspar, the material becomes progressively more brittle and stronger. Studies on the frictional resistance to sliding have shown that a decrease in frictional strength only begins when the amount of clay approaches 25-30% of the rock volume [15]. As the percentage of clay increases the frictional strength decreases monotonically until the amount reaches 70-75%, after which little change occurs. Studies of granitic rocks have found similar changes when the percentage of phyllosilicates is varied [20].

A second parameter is the nature of the phyllosilicate species. Illite and kaolinite have been found to have much less affect in altering the strength of rocks than do species containing interlayer water such as the smectites. This is found in both the frictional strength as well as in the mechanical behavior of samples that are 100% clay [21].

Mechanical behavior common to many shales as illustrated in tests on samples that are composed of: quartz plus feldspar 37-45%, illite/mica 27-31%, kaolinite 10-17% and chlorite 10-16%. Specimens were deformed in triaxial compression at constant strain rates of 10⁻⁵ and 10⁻⁷s⁻¹, at confining and pore pressures to 200 MPa and temperatures to 225° C. In experiments done at room dry conditions and confining pressures to 200 MPa, a transition from brittle to semi-brittle behavior is observed, with all specimens eventually failing in shear fractures. When compared to other common sedimentary rocks the strengths are not significantly different (Figure 3); they are generally stronger than the highly porous limestones, but weaker than some marbles. They are weaker than Berea sandstone containing 12% porosity at low pressures, but are about as strong at higher pressures. The
significant difference is that at 150 MPa confining pressure, other rocks have become ductile, while even at 250 MPa, the shale, although deforming with shear bands, eventually fails along shear fractures. This semi-brittle behavior is characteristic of dolomites and low porosity sandstones under similar conditions.

When experimental temperatures are raised, specimens still in a room-dry state show about 40% reduction in strength from 55° to 190° C and the deformation is still semi-brittle at the higher temperature (Figure 4). In a series of tests under saturated conditions using distilled water at room temperature with constant pore pressures ranging from 0 to 70 MPa, with confining pressures of 70 or 90 MPa, all tests produced shear failure as in dry specimens.

When specimens are deformed saturated and with controlled pore pressures, at room temperature the strength is about 17% of that when specimens are deformed in a room dry condition. Under elevated temperatures the strength does not change significantly at temperatures to 225°C; apparently the presence of pore water is the primary control on strength, with temperature having minimal influence over this range.

The importance of the pore fluid is further demonstrated in the experiments where the pore pressure is controlled throughout the test. It is found that the concept of effective stress, where the mechanical response is a function of the difference in confining and pore pressure and not the absolute value of either [22], does not hold for these tests. Here the rocks respond to the absolute value of pore pressure with strength varying inversely as the pore pressure is increased even at the lowest strain rate investigated. The latter, 10⁻⁷s⁻¹, is sufficiently slow to allow pore pressure equilibration even in igneous and metamorphic rocks. This effect has also been reported for Pierre shale where permeabilities were measured to be 10⁻¹⁵ to 10⁻¹⁶ m² [23].

SEM observations of undeformed specimens show grains of quartz and feldspar interleaved between clay plates that have a dimensional preferred orientation parallel to bedding. This texture appears little altered in specimens deformed at temperatures to 190° C and pore pressures to 20 MPa except along the shear fractures. As in the mechanical response, the differences occur between specimens deformed under dry and those in saturated conditions. In both cases the fracture surfaces are generally planar and well defined macroscopically. Under SEM, both show grooves parallel to the shear displacement and both show some granulation of the material along the fracture. But the degree of comminution of the dry material is much higher than that of the saturated shale and the damage zone is much more extensive. Along the surface in the saturated specimens the clays are re-oriented so as to parallel the shear displacement. Here the rock is highly indurated, more so than the material away from fractures. This zone is a few millimeters wide but introduces an anisotropic fabric to the fracture zone. The result is that the fracture surface itself is smoother and more planar. The re-orientation of the clays along the fractures suggests that permeability may be reduced at high angles to the fracture.

Argillaceous Rocks and Fluid Flow Along Faults

To what extent do the mechanical properties of the shale alter the fluid transport properties of the faults? Experimental studies of the frictional properties of clays have shown that the same fabric geometries described above are present but the orientations are often defined by orientation of clay particles rather than fractures [15]. In such experiments Y-fractures do develop with sufficient shear strain. Fluid flow along them is potentially viable, however other fabric elements are often not manifested as fractures. In field studies where the country rock is shale and the fault rock a shale cataclasite, Y-fractures have served as avenues for multiple cycles of fluid migration [24]. In such cases, although flow across the faults appears to be inhibited, the faults are still acting as conduits, despite the presence of shales adjacent to and within the fault.
In contrast, the presence of shales has often been interpreted as evidence for sealing faults both across and along their trace because of the presumed ductile nature of the rock [25, 26]. Such cases have been related to “shale smears” along faults. The latter have been best documented in growth faults where faults cut interbedded sandstones and shale sequences and where the shales are not highly indurated [27, 28]. These studies have also concentrated on situations where the fault displacement and related shear strain is relatively small with respect to the bed thicknesses. Additional attempts have been made on the basis of experimental and field observations to quantify the potential of shales to form seals in terms of “clay smear potential” or “shale smear factor” for a given sedimentary sequence [29]. Such efforts have been largely empirical and without quantification of permeability measurements. Research is necessary to extend these observations to shales that are more indurated.

Briefly, the foregoing can be summarized:

- Shales vary widely in composition but younger ones commonly contain about 40-45% quartz, feldspar, carbonates or other framework minerals and about the same amounts of phyllosilicates.
- When nominally dry their mechanical properties are not significantly different from other rock types, it only when saturated that their strength decreases dramatically, as does their shear resistance.
- The presence of water appears to suppress significant sensitivity of strength and a transition to ductile behavior to increases in temperature.
- In contrast to most other rocks, over a wide range of strain rates, shales do not behave in accordance to the concept of effective stress.
- Within a shear fracture or fault, displacement re-aligns clays parallel to the slip direction creating a potential barrier to fluid flow across the failure surface.
- Smear fabrics in small-displacement faults have been recognized in both laboratory experiments and field studies in interbedded shale and sandstone sequences where the shale is poorly lithified; the ability to predict such behavior still requires additional study.

**Fluid Chemistry, Fault Rock Fabric and Fluid Flow**

Although the presence of fluids and the potential for elevated fluid pressures associated with faults has been discussed since the seminal paper by Hubbert and Ruby, [30] the questions of fluid-rock chemical reactions have only been addressed relatively recently. Chemical reactions, especially those which would alter minerals to phyllosilicates have been envisioned to reduce fault strength and permeability. Solution-transport and deposition of material within faults, or fault healing, has been suggested as a method of increasing fault strength and reducing permeability. But such processes also indicate the possibility of increasing the brittle response resulting in fractures or faults of increased permeability. While a few recent experimental and field studies have started to assess the physical processes involved [31-33], great uncertainty remains about the implications for fault fabric and geometry and subsequent fluid migration.

Experimental studies on sandstones that contain 8-10% clays show that chemically active solutions such as NaCl may reduce the frictional strength by as much as 45% [33]. This reduction is apparently the result of chemical interactions between the clays and quartz and localized in the vicinity of the clay concentrations. Depending upon the strength of the fluids either fracturing of quartz is enhanced to produce gouge reducing the frictional resistance, or the quartz is dissolved and removed decreasing the contact area. The importance of clays in facilitating chemical interaction has been found in other experimental studies of fault gouges [34-36].
Recent field studies have begun to document chemical changes that take place as a result of fluid migration through fault zones where shales are involved [9, 24]. One such study analyzed displacement and fluid migration in and adjacent to normal faults along the coast of the Bristol Channel, UK [24]. The faults cut a sequence of limestones and shales which show both normal and reverse displacement with offsets of tens of meters. Shale samples taken at distances up to 20 m away from the faults show average compositions of about 16% quartz and feldspar, 31% calcite, 3% kaolinite, 3% chlorite, 33% illite and 2% smectite. Samples analyzed from within eight faults have an average of 23% quartz and feldspar, 45% calcite plus dolomite, 5% illite and no significant changes in the other clay species. These data indicate significant enrichment of carbonate coincident with marked removal of illite as a result of fluid migration through the fault rocks. Geochemical samples show the fluids were restricted from less than 0.5 m up to 4 m on either side of the faults. Fluid inclusion studies show that at least three generations of fluid migration took place in some faults. The influence of the fluids on mechanical behavior is shown by a strong correlation of the width of the mineralized zone with displacement, suggesting that the fluids decreased the frictional resistance to slip.

Such studies suggest that dissolution and removal of material is equally important as deposition of minerals by advecting fluids in altering the fault properties. It is clear, however, that much more research needs to be undertaken before the importance of such effects can be assessed.

**Future Research Directions**

Presently, we have established a basis to evaluate the capacity of fault zones in general for fluid flow, however, criteria to quantify the potential of argillaceous rocks and faults that cut them to act either as conduits or seals currently needs more research. The mechanical and physical properties even of intact argillaceous rocks remains poorly defined especially over repository time frames. Of possibly more importance, healing processes and their kinetics are pivotal to any prediction of life of a repository. It is necessary to develop scaling relationships for permeability involving single phase flow and migration of multiphases. Fundamental is the necessity of relating laboratory measurements to field conditions, and using field measurements to constrain laboratory tests. Potential modifications of flow and sealing intervals with time resulting from changes of state of stress, fluid compositions, and boundary conditions need to be studied. The nature and kinetics of geochemical interaction between fluids and rocks under non-hydrostatic stress fields have only begun to be explored and yet are critical to the problems at hand. Finally, development of diagnostic features that allow recognition of potential of lithologies, faults and fractures for fluid flow or sealing are desirable if we are to continue beyond an empirical base. Clearly, such research is multidisciplinary requiring geological, geophysical, geochemical, experimental, field and theoretical collaboration.

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Figure 1. Schematic diagram of geometry and relative properties of an idealized fault zone. The surfaces show general variation in properties along and across fault. Vertical arrows show increasing relative values of respective properties.

Figure 2. Fracture array showing relative geometries defined by triaxial compression experiments on simulated fault gouge.
Figure 3. A comparison of ultimate strength versus confining pressure for this shale with other rocks commonly tested in the laboratory. All specimens were deformed room dry.

Figure 4. Summary plot of experiments on shale showing differential stress at ultimate strength versus temperature for tests done at an effective confining pressure of 50 MPa.
SESSION II

Flow in Faults and Fractures
Chairmen: A. Gautschi (NAGRA, Switzerland), and P. Lalieux (NEA)
The Impact of the Earthquake Cycle on Fluid Flow Through Fractures in Argillaceous Formations

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Abstract

Employing empirical observations of the hydrological changes that follow major earthquakes it becomes possible to predict subsurface fluid-flows that accompany active tectonics. Levels of coseismic strain and the consequential hydrogeological changes, are largest close to jogs in the plane of fault-rupture. Low permeability argillaceous formations may be either passively or actively affected by the hydrogeological changes that accompany coseismic strain-cycling. Fluid release is associated with consolidation of the sediment at greater depth, as well as with coseismic strain, and this in turn can generate intra-formational faulting or the remobilisation of the argillite and the eruption of mud volcanoes, sometimes explosively. In areas of active tectonics, intermittent hydraulic activity along fractures may be a feature of low permeability sedimentary formations overlying confined aquifers.
Introduction

In a series of earlier studies, hydrological changes accompanying numerous earthquakes in many different parts of the world were collected, analysed and, where possible, quantified (Muir Wood & King, 1993). In order to resolve such changes it is first necessary to focus on areas where there is hydrogeological communication from the surface to depth. Such communication is most obvious where there are high permeability deep-sourced conduits, releasing thermal springs, but has also been demonstrated more widely from certain types of reservoir-induced seismicity in which earthquakes are triggered by a raised fluid pressure at the surface. (Focussing on areas in which there is no barrier between surface water and fracture flow deep in the upper crust inevitably disqualifies the collection of observations from sedimentary basins.)

Within this study it was found that once coseismic hydrological effects could be isolated from an inevitable groundwater 'noise', a simple relationship could be demonstrated between the style of fault displacement and its spatial hydrological 'signature' (Muir Wood and King, 1993). In most igneous and metamorphic rock, as well as well-lithified sediments, mobile water is held and transported in fractures. The response of these fracture systems to the coseismic strain field of different classes of earthquakes, indicates that porosity is responding to the prevailing stress-state, as is implied by numerous experimental studies (Walsh, 1965, Batzle et al., 1980). In a region undergoing extensional faulting, continuing strain distributed through the crust causes appropriately oriented high-angle fractures to dilate, thereby increasing crustal porosity. Pore-pressures are sustained by slow infilling of the dilated fractures with fluid. At the time of a major normal fault rupture, strain, formerly distributed through the crust, becomes concentrated on the fault. As a result the region of the crust that 'surrounds' the fault (ie. the footwall and hangingwall) undergoes elastic rebound in compression. In contrast, in a region undergoing compressional tectonic deformation, in the interseismic period negative strain (volume decrease) in the crust closes high angle fractures and reduces crustal porosity. At the time of the fault rupture, as strain is transferred into fault displacement the surrounding crust undergoes elastic rebound in extension.

Hence in the region around a normal fault rupture the coseismic decrease in crustal porosity should lead to a regional postseismic expulsion of water. In the area surrounding a compressional fault rupture coseismic porosity expansion reduces hydraulic pressures leading to water being drawn into the crust. In areas of low precipitation river flows provide the most important resource for quantifying these post-seismic changes. Following the M7.5 1959 Hebgen Lake normal fault earthquake the cumulative volume of water discharged was equivalent to ca. 0.5 km$^2$ with typical decay times to half peak flow of 100-150 days. The volumes and extent of water release are consistent with the levels of modelled coseismic strain (Muir Wood and King, 1993).

Close to the fault, irregularities in fault geometry, and variations in displacement, begin to dominate the strain-field. The most notable effects accompany jogs where displacement passes from one fault plane to a parallel one (see Sibson, 1986). As jogs can have a dimension of metres to kilometres, and for major earthquakes involve the transfer of several metres of displacement, self-evidently they can be subject to very significant levels of strain; potentially higher than $10^2$. This strain can be either compressional or dilational. Within the jog the coseismic stress changes do not simply accompany elastic rebound but instead are loaded directly by adjacent fault rupture. Along some strike-slip and oblique-slip fault ruptures, where jogs communicate with the surface, these effects may cause spectacular coseismic fountain outbursts at pressures of several bars (as in the October 28, 1983 earthquake at Borah Peak, Idaho, Waag & Lane, 1985) or a significant collapse of the water-table (as
along parts of the Fairview Valley, Nevada fault in the 16th December 1954 earthquake, Zones, 1957).

**Implications for Low Permeability Argillaceous Formations**

These empirical findings have a number of important implications for the understanding of the potential for tectonically induced fluid flows in low permeability argillaceous formations. Direct observations from such formations are inevitably scarce, as they are not a target for fluid exploitation.

A simple distinction is worth pursuing between “active” hydrogeological changes associated with a formation that is itself subject to strain, and “passive” changes that result from an external increase in fluid pressures. Inevitably there are situations in which both active and passive behaviours may combine.

**Strain Cycling of Argillaceous Formations**

Post-seismic observations of hydrology reveal that strain through the seismic-cycle must chiefly be accommodated in rocks of the upper crust by changes in the aperture of fractures. This strain originates in tectonic boundary forces and becomes relieved through earthquake fault displacement. The degree to which a shallowly buried geological formation currently participates in such strain-cycling may be determined by the variation of horizontal tectonic stress with depth through the formation, that will reflect its connection to the underlying crust and its ability to behave elastically. Some shallow formations may, as a result of underlying detachment or internal creep, not respond elastically.

Where a formation participates in the accumulation and release of tectonic stress through the earthquake cycle, this will require strain-cycling, that in the region around a major fault implies strains of the order of $10^3$. In a crystalline rock this strain is associated with the dilation and closure of fractures, and this is also likely to hold true in a well-consolidated argillaceous sediment. The impact such strain has on formation permeability will be determined by the pre-existing crack population, and the way in which strain is concentrated on a few specific larger fractures or distributed over a broad microfracture population. Some observations from crystalline rocks suggest that strain concentration on larger fractures is to be expected (Muir Wood et al., 1996), although there will be less tendency for such concentration where the formation is of low rigidity.

Strain changes, accommodated on fractures, inevitably imply a change in formation porosity. In very low permeability argillaceous formations, porosity changes will result in modified fluid pressures. Positive, volume increasing strain (as is found inter-seismically around a normal fault, and post-seismically around a reverse fault) will lead to under-pressureisation, just as volume decreasing strain (as is found inter-seismically around a reverse fault, and post-seismically around a normal fault) will lead to over-pressureisation. The magnitude of the effect will be dependent on the original porosity of the sediment, the lower the porosity: the more magnified the effect. Permeability is itself likely to be determined by the population of microfractures, and consequently argillaceous sediments that show the most significant strain-induced changes in pore fluid pressure are likely to take the longest time to re-equilibrate. In fact it would be more likely that the rock would remain a closed system throughout the seismic strain cycle, its internal fluid pressures acting like an internal aneroid barometer to strain. Clearly if fluid pressures at some part of the cycle exceed those of the overlying lithostatic load, localised hydrofracture will result.
Strain may also result from non-tectonic processes. Over-pressures can result from an increase in surface load resulting from sediment or ice-sheet accumulation while underpressures can reflect erosion or glacial unloading (or even downslope processes in areas of high topography).

Zones of overpressurised argillaceous formations are commonly encountered in hydrocarbon exploration in areas of rapid sedimentation, as along the Gulf Coast of the USA. In certain circumstances over-pressurisation can drive its own localised intra-formational "tectonic" processes. During the Cenozoic the North Sea rifted basin became infilled with up to 3 km of chiefly argillaceous sediments. Over wide areas these sediments contain two tiers of internal faults, typically 500 m-1000 m long, 300 m high and spaced 200-500 m apart (Cartwright, 1994). The faults, dipping between 30° and vertical, have displacements of up to 100 m and are organised into giant polygons. Equivalents of these structures are also seen in onshore exposures of the Palaeogene argillaceous formations in East Anglia and Belgium (Henriet et al., 1991), where displacement can be shown to decrease downwards.

The overall pattern of the faults suggests that they were initiated around domes developed in the uncompacted argillaceous sediments during rapid burial and loading. The faults themselves were both triggered by high effective fluid pressures and, like mudcracks, acted as conduits for flow to relieve these pressures. To accumulate displacements of up to 100 m, displacement and fluid pressure release must have been repeated on many occasions. The widespread distribution of pockmarks in the North Sea floor suggests that some of these intra-formational faults may continue to be active as conduits for fluid flow.

The Passive Response of Argillaceous Formations

From the pattern of coseismic surface fluid flows found around faults in crystalline crust, it is possible to conceive the changes in fluid fluxes at the base of an argillaceous formation overlying a region of active extensional tectonics. In general a low permeability formation will cause such flows to be diverted laterally across a sedimentary basin. However it is always possible that there are isolated fault-related conduits of high conductivity passing through the formation.

Mud-volcanoes are an important manifestation of the existence of highly conductive apertures associated with argillaceous formations (and also of the ability of argillaceous formations to re-liquefy). Most mud volcanoes are found in regions of rapid loading and compressional tectonics. The mobilisation of the argillite requires a concentration of fluid, either arriving from deeper levels in the crust, or from within the formation itself. Most mud volcanoes, like thermal springs, are probably fault controlled, being located at dilational jogs along strike-slip faults (that may in turn be minor transfer faults of the dominant reverse fault structures). Mud volcanoes can also cluster along the extensional axes of anticlines formed above blind reverse faults. Clusters of mud-volcanoes sometimes show alignments reflecting this tectonic influence.

There are many observations of mud volcanoes erupting coseismically (in major earthquakes in Burma and Iran for example) as a result of the strain changes accompanying faulting. However interseismic strain can also trigger a build up of fluid pressures sufficient to cause an eruption, as has been witnessed in areas like the Arakan coast of Burma and in southern Sakhalin. On March 20th 1959 one mud volcano around the shores of Terpeniya Bay (Sakhalin) erupted 200,000 m³ of Cretaceous mud and rocks (and entire trees!) to a height of more than 100 m, over an area of 0.6 km² (Gorkun and Siryk, 1968). Elevated subsurface fluid pressures associated with a nearby retreating ice-
sheet can also cause a pressurised outburst, as evidenced at the 300 m diameter Howe Lake, hydrodynamic blow-out crater in southeast Saskatchewan (Christiansen et al., 1982).

Conclusions

Seismic strain-cycling has the potential to change the hydrogeology of low permeability argillaceous materials, first by affecting the apertures of microcracks distributed through the formation and second by imposing altered fluid pressures at the boundaries, or within, the formation. In a medium where porosity is dominated by fractures (or connected with such a medium) sealed low permeability reservoirs have the potential to act as strain-barometers recording changes in dynamic porosity through an alteration in fluid pressure (Bodvarsson, 1970). Coseismic and interseismic strain changes may explain the development of some zones of over- and under-pressurisation within confined fluid reservoirs, where other explanations associated with consolidation, secondary porosity reduction or temperature changes are found to be insufficient (Domenico and Palciauskas, 1988). The impact on pore-fluid pressures will be determined by the dynamic porosity relative to the strain-sensitivity of the fractured medium.

The presence of potentially conductive apertures passing through low permeability formations, reveals the difficulty of attempting to sample and scale the permeability of a formation from local observations. On a large scale, formation permeability may be dominated by the behaviour of a small number of apertures, and in the case of mud volcanoes and other fluid outbursts associated with internal fault displacement (as in the Palaeogene sediments in and around the North Sea), high permeability may be very transient. Where an argillaceous unit is to be considered for the long-term disposal of radioactive wastes, formation permeability may need to be considered in terms of its potential variability in both space and time. Even where crystalline rocks are buried beneath a relatively low permeability sedimentary cover a moderate sized earthquake can have a significant transient effect on near-surface hydrology (Somville, 1939).

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References


Evidence for thresholds, pathways and intermittent flow in argillaceous rocks

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Abstract

With matrix hydraulic conductivities in the range $10^{-12} - 10^{-15}$ m.s$^{-1}$, beds of compact mudrock should represent very significant barriers to fluid migration. However, if the forces (fluid potential gradients) which drive fluid flow in the geological environment, rise to the point where they exceed the flow resistance of these barrier formations, then fluids will migrate through the mudrock by exploiting "preferential pathways" which can be considered to be pathways of least resistance. We can refer to the situation in which flow is actually imposed on a low permeability formation as one of "forced-advection". In many cases, the incipient pathways of fluid flow are already present as faults, transecting the low permeability formations, or as interconnecting networks of smaller fractures or fissures. Under special circumstances, it would appear to be the case that very high fluid pressures can actually create the pathways of fluid migration through mudrock formations.

The authors explore the effects of stress, stress history and current depth of burial on pathway flow in mudrocks and propose that a self-sealing mechanism operates at depths exceeding some critical value, specific to each rock type. The effective stress acting at the critical depth may be related to the so-called "critical state" of theoretical soil mechanics. Mudrocks at depths less than critical are susceptible to brittle fracturing and associated shear-dilation, suggesting that pathways will be open (i.e. dilated) and capable of conducting fluids. Mudrocks at depths greater than critical will tend towards a more plastic mode of deformation and will compact when sheared. A self-sealing pathway in a deeply-buried mudrock will exhibit a well-defined threshold pressure for flow. Above this threshold, the permeability of the pathway will exhibit a highly nonlinear dependency on local fluid pressure. It is possible for a system with these characteristics to show intermittent or episodic behaviour, particularly when the mobile fluid is a gas. A number of case histories are examined and used to illustrate some of the more important processes affecting fluid movement in argillaceous formations.

Introduction

Diagenesis is the process by which sediments become lithified during burial and the extent of diagenetic alteration is, perhaps, the most significant factor in determining the physical, chemical and hydrogeological characteristics of a mudrock. Diagenesis is primarily a response
to the changes in temperature, stress, fluid pressure and chemical environment which are occasioned by burial. Some of the principal diagenetic processes occurring during burial are: (a) compaction and fluid migration, (b) development of diagenetic bonds, (c) mineralization and the introduction of interparticle cements, (d) organic reactions, (e) clay mineral dehydration and transformation, and (f) pressure-solutioning and recrystallization. Eventually burial diagenesis gives way to metamorphism, but there is no clear boundary between these two realms.

The character and properties of many mudrocks are not exclusively determined by burial diagenesis, since a number of processes occurring subsequent to burial can play an important part in determining the structural attributes of these rocks. The most important of these processes are tectonic deformation, uplift and exhumation and, not infrequently, all three processes are closely interrelated as surface erosion strips away sediments thrown up by large-scale deformation, bringing the mudrock stratum closer to the surface. The combined effect is to impose a stress history on the mudrock and it is this history that is the second major modifier of the character and properties of many argillaceous rocks (Horsemann and Volckaert, 1995).

**Stress history of a mudrock**

Figure 1 provides a simple picture of some of the main effects of burial and subsequent exhumation on a stratum of mudrock (Skempton, 1964; Fleming et al., 1970). At the time of deposition, shown as point (a), a mud may have a water content in the range 80 - 100%. As additional sediments accumulate on top, their weight must be borne by the mud layer. The increase in stress causes water to be expelled leading to a decrease in porosity and void ratio. Provided that the deposition of sediments is not interrupted and is not too rapid, the newly-formed mudrock will follow the path defined by points (a), (b) and (c). The fabric will alter in response to the increased effective stress and the platy clay minerals will become orientated at right angles to the vertical compression direction. Using soil mechanics terminology, the mudrock at any point on this path is *normally-consolidated*, since its porosity and water content are always commensurate with its depth of burial. A mudrock in this condition is considered to be in a state of hydraulic equilibrium with its surroundings.

If deposition is extremely rapid, it may not be possible for excess pore pressures developed in the early stages of compaction to fully dissipate over the available time scale. The mudrock will then exhibit an overpressure and its porosity, void ratio and water content will each be higher than might be anticipated from its current burial depth. The mudrock is then sometimes referred to as being *underconsolidated* or undercompacted. A mudrock in this condition is considered to be in a state of hydraulic disequilibrium.

At point (c) deposition ceases. If the depositional phase is immediately followed by a period of erosion, then the mudrock follows path (c) to (d). The material is subject to an effective stress which is less than the maximum burial stress and is said to be *overconsolidated*. The reduction in effective stress is accompanied by an increase in water content (known as rebound), but this increase is far less than the decrease in water content during consolidation. Thus, although the material at point (d) is under the same effective stress as the material at point (b), the water content of the overconsolidated mudrock is considerably less. The particles are therefore in a much denser state of packing than the normally-consolidated mudrock.
Figure 1 - Schematic of the effects of a simple cycle of deposition, burial, erosion and exhumation on the water content of a mudrock and on the state of stress in the mudrock stratum. Mudrock on paths between points (c) and (d) or (c') and (d') is defined as “overconsolidated” (modified from Skempton, 1964 and Fleming et al., 1970).

If there is a considerable time-lapse between the end of the depositional phase and the start of erosion then the mudrock may follow the path (c) to (c') which is characterized by a loss in water content under constant effective stress and by the development of diagenetic bonds. This period may be sufficiently long for minerals to recrystallize, for strong particle-particle adhesion to develop, or for a cementing medium to be precipitated in the interparticle spaces. The more strongly developed the interparticle bonding, the smaller will be the increase in water content on unloading along path (c') to (d').

Figure 1 also shows the relationship of the horizontal effective stress, $\sigma_h'$, to the vertical effective stress, $\sigma_v'$, during the deposition/erosion cycle. The ratio $\sigma_h' / \sigma_v'$ is known as the coefficient of earth pressure at rest, $K_0$. The value of $K_0$ must lie within upper and lower bounds which are determined by the long-term strength properties of the mudrock. In effect, if $K_0$ is excessively large or small, the shear stress will exceed the strength leading to plastic deformation. During the depositional phase, $K_0$ (n.c.) for the normally-consolidated mudrock is generally found to lie in the range 0.4 to 0.7.

During erosion, the vertical effective stress will decrease in a manner which is in line with the reduction of overburden thickness. However, in order for the horizontal effective
stress to decrease, the stratum must be able to accommodate horizontal strain. Since this is restricted by the lateral continuity of the layer, the horizontal stress may remain high, while the vertical stress decreases (Skempton, 1961). This results in a gradual increase in the coefficient of earth pressure at rest as the mudrock moves along path (c) to (d). Typical values of $K_0$ (o.c.) for an overconsolidated mudrock are generally in the range 1 to 4, with the maximum value probably limited by shear strength. The state of stress developed in a highly-overconsolidated mudrock at shallow depth can therefore lead to spontaneous shear failure, dilation, and the formation of small reverse faults.

![Graph showing hydraulic conductivity vs. depth and effective stress](image)

**Figure 2** - Depth trends in hydraulic conductivity for the Cretaceous shales of S. Dakota (after Neuzil et al., 1984; reproduced with permission). The disparity between laboratory and field testing results and the values estimated by hydrogeological modelling suggests that cross-flow may be focused along widely-spaced fractures. The depth trends suggest that these pathways may be closed below the critical depth of 2200 m.

**Fracture flow, stress-sensitivity and critical depth**

Based on very extensive studies of the Cretaceous shales of S. Dakota, Neuzil et al. (1984) examine the possibility that cross-flow or “leakage” through these shales might be focused along fractures. The groundwater flow system in S. Dakota is dominated by three major aquifers and their confining layers: the limestones of the Madison Group, the sandstones of the Inyan Kara Group and the Dakota-Newcastle sandstone. Surface recharge to the aquifers occurs at outcrops in the Black Hills and discharge occurs at the Dakota sandstone outcrop in the east; groundwater flow is predominantly from west to east. The confining layer overlying the Dakota aquifer consists of three Cretaceous shales: the Mowry-Belle Fourche, the Carlisle and
the Pierre. The numerical simulations of Bredehoeft et al. (1983) had demonstrated that conditions in the Dakota aquifer, prior to the development of water resources, could only be explained by a finite rate of upward leakage through these overlying shales.

Data on hydraulic conductivity and specific storage of these shales was obtained using three techniques: (1) numerical simulations of the regional groundwater flow system, (2) *in situ* field tests, and (3) laboratory tests on core samples. Two types of *in situ* tests provided hydraulic conductivity data for the shales: slug tests conducted in a 180 m borehole in the Pierre Shale and a pumping test in the Dakota sandstone which had been performed some years earlier. The data from the pumping test was re-analysed using leaky-aquifer theory to provide a value for the product of hydraulic conductivity and specific storage of the shale. Cores extracted from several boreholes were tested in the laboratory using pulse decay and consolidation techniques. The laboratory and field measurements of hydraulic conductivity both showed a consistent decrease with effective stress and depth (Figure 2). A conspicuous feature of this plot is the very large discrepancy (1 - 3 orders of magnitude) between the measured values and those inferred from the regional modelling exercise.

Neuzil et al. (1984) suggest that this discrepancy can be explained by vertical flow along a system of fairly widely-spaced fractures. Using a simple parallel-plate model for fracture flow, the authors performed a number of scoping calculations to examine the interrelationships between spacing, aperture and flow. The observations might be explained by a system of fractures with spacings in the range 0.1 - 1 km. The decrease in estimated regional conductivity in successively deeper shales might then be interpreted in terms of fracture aperture reduction under increasing compressive stress.

Extrapolation of the depth-trends reported by these authors leads to the important new observation that the trend for local-scale (matrix) hydraulic conductivity converges with the regional trend at a depth of around 2200 m. Although this is well below the base of the deepest formation, it suggests that the fractures responsible for the scale-dependency would be closed at this depth. The effective stress acting at this *critical depth* would then represent the closure stress of the flow pathways.

**Simple conceptual model of pathway flow**

Figure 3 shows a simple conceptual model of pathway flow in a mudrock. A source reservoir is linked to a sink reservoir by a flow pathway which is assumed to be vertical fracture with a variable local hydraulic aperture denoted by $\delta$. Taking the simplest case of a smooth-walled planar fracture, the flow of fluid per unit width of the fracture can be quantified using the cubic law:

$$Q = -\frac{\delta^3}{12\eta} \left(\frac{dp}{dz}\right)$$  \hspace{1cm} (1)

where $\eta$ is the dynamic viscosity of the fluid and $dp/dz$ is the pressure gradient of the fluid along the pathway. The horizontal (total) stress, $\sigma_h$, acting normal to the plane of the pathway must
be transmitted, in full, across the flow pathway. It is assumed that the local hydraulic aperture of
the pathway shows a functional dependence on the difference between the horizontal stress
and the local fluid pressure:

\[ \delta = f(\sigma_h - p) \quad (2) \]

Although this functional relationship might be quantified in terms of the elastic
properties of the mudrock, the role of inelastic deformation (e.g. plastic and/or time-dependent
mechanisms) in determining pathway aperture demands very close attention.

![Diagram of pathway flow](image)

**Figure 3** - Simple conceptual model of pathway flow in a mudrock. A source reservoir is linked to
a sink reservoir by a vertical preferential pathway which is assumed to be a planar fracture with
a stress-dependent local hydraulic aperture.

Building on the above hypothesis, we might suppose that there is some critical value of
the quantity \((\sigma_h - p)\) which leads to total closure of the flow pathway (i.e. \(\delta = 0\)). The **sealing
criterion**: can then be written:

\[ Q = 0 \quad \text{for} \quad (\sigma_h - p) > (\sigma_h - p)_{\text{crit}} \quad (3) \]

As fluid pressure gradually builds up in the source reservoir, there will come a point at
which the sealing criterion is no longer met and fluid will start to flow towards the sink. The
introduction of the pathway aperture relationship of (2) into the cubic law of (1) makes the
effective permeability of the pathway a highly nonlinear function of the local fluid pressure.
The resulting flow law therefore exhibits both a **threshold** and a **nonlinearity**. An increase in
fluid pressure in the sink reservoir above the critical pressure will therefore lead to dilation of the
flow pathway and a substantial increase in effective permeability. The case for the existence of a

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1 It is important to note that fluid pressure \(p\) is not necessarily equal to the pore pressure in the surrounding mudrock
since the formation may not be in a state of hydraulic equilibrium.
well-defined threshold is actually strongest where the fluid is a gas or a liquid hydrocarbon (i.e. an immiscible phase in water), since the passage of such fluids through very narrow intergranular spaces is opposed by the forces of capillarity.

Sealing mechanisms

The representation of a flow pathway in a mudrock as a smooth-walled fracture is obviously an oversimplification. A conspicuous difficulty with the simple conceptual model is that the normal stress acting across a smooth-walled fracture of finite aperture could never exceed the fluid pressure within the fracture. In order for a fracture to transmit a normal stress which exceeds fluid pressure, it must have rough surfaces so that the stress can be transmitted via contacting asperities, or it must contain an infilling or fault gouge which is, itself, stress-supporting. Although the simple cubic law of (1) might no longer quantify the movement of fluid within such a pathway, we might still expect to see a sensitivity of pathway permeability to applied stress and a resulting nonlinearity in the flow behaviour.

![Diagram](image)

**Figure 4** - Example of a shale seal separating reservoir sandstones in the Tuscaloosa Formation of Louisiana (after Engelder et al., 1994). The lower reservoir is overpressured. The 20 m bed of shale supports a pressure gradient of 0.43 MPa per metre. At this depth the shale is likely to be plastic, leading to self-sealing pathways. The horizontal stress acting in the shale probable sets the upper limit for the overpressuring of the lower reservoir.

Potential fracture sealing mechanisms in mudrocks include: (a) plastic yielding of stress-supporting asperities, (b) compaction of a clay-rich fault gouge under high effective stresses, (c) pressure-solutioning and re-precipitation of fracture-infilling minerals, (d) swelling of the disrupted fabric of the wallrock, and (d) precipitation of minerals from flowing
groundwater. The majority of these mechanisms are stress-dependent. The petroleum industry classify shale caprock seals as pressure seals and capillary seals. Typically, the pressure seal is a bed of overpressured and underconsolidated shale overlying the reservoir. It is supposed that the very high porewater pressures and adverse pressure gradients acting within this bed totally inhibit the passage of brine, gas and oil through the seal. The capillary seal on the other hand is effective only for gas and oil and, as the name suggests, the seal is considered to be effective because the very fine pores of the shale lead to exceptionally high capillary entry pressures for hydrocarbons. A bed of overpressured shale might act as both a pressure seal and a capillary seal (Magara, 1978).

**Some generalities in material behaviour**

One approach to quantifying the deformation, failure and pore pressure responses of granular soil-like media involves the use of elastoplastic models based on the concept of the critical state (Schofield and Wroth, 1968; Atkinson and Bransby, 1978; Wood, 1992). The underlying idea of these models is that it possible to define a stress state for the material in which very large shear deformation produces no change in pore volume or pore pressure. This is known as the critical state (Roscoe and Burland, 1968).

If we examine the same material over a range of water contents, the locus of all possible critical states in void ratio - effective stress space\(^2\) is known as the critical state line (CSL). On one side of the CSL, known as the “wet side”, the material will compact (or consolidate) during shear deformation. On the other side of the line, known as the “dry side”, it will dilate during shear. When applied to cohesionless soils (e.g. sand), these mechanical responses are interpreted in terms of shear-induced changes in the packing arrangement of the particles. Although there is no clear explanation of such responses in clay-rich media, it would appear that the tendency to compact or dilate is strongly dependent on the particle-particle physico-chemical forces, with the forces responsible for the adhesion of clay platelets emerging as particularly significant to dilation. Since the magnitude of these forces will depend, in part, on the maximum burial depth, they are likely to exercise an important control on the strength and volumetric behaviour of an overconsolidated clay (Horseman et al., 1996).

Given that critical state concepts were developed to describe the mechanical responses of “soil-like” materials, we might ask whether an approach based on these concepts has any validity when applied to rocks such as shale? All rocks exhibit shear-dilatant behaviour when loaded under zero or low levels of confining pressure, associated with the initiation and propagation of microcracks (Brace, 1978). As the failure stress is approached, these microcracks coalesce to form the macroscopic shear planes characteristic of brittle failure in unconfined compression. As confining pressure is increased, the stress levels necessary to cause crack propagation also increase, leading to an overall strengthening of the rock. The application of a large confining pressure can lead to the partial or complete suppression of dilatant brittle fracturing, with plastic (or viscoplastic) deformation emerging as the dominant mechanism at very high confining pressures (Evans et al., 1990). This change in mechanical behaviour is gradual and is known as the brittle-ductile transition (Heard, 1960; Griggs and

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\(^2\)The critical state line is usually represented in three-dimensional space where the third dimension is the shear stress. Strict terminology would identify the delimiting line in void ratio - effective stress space as the projection of the critical state line.
Handin, 1960). Once in the fully-plastic state, many rocks are capable of undergoing isovolumetric flow (i.e. flow at constant volume). The application of even higher levels of confining pressure then leads to little additional strengthening (Evans et al., 1990). However, if the rocks retain some porosity when they enter the fully-plastic state, they can continue to compact (or consolidate) under increasing levels of effective confinement, leading to further strengthening of the fabric.

**Figure 5** - Void ratio (or porosity) versus mean normal effective stress for a mudrock undergoing normal burial and compaction followed by uplift, erosion and exhumation. The critical state line (CSL) divides the overconsolidated domain into two regions, the “dry side” to the left where the material tends to dilate and the “wet side” to the right where compaction (or consolidation) is favoured.

In most rocks, the brittle-ductile transition occurs at high levels of confining pressure and at the elevated temperatures typical of moderately deep crustal conditions. Mudrocks are exceptional in their behaviour and true plasticity can usually be attained at earth surface temperatures and relatively modest levels of effective confining pressure. The very fine-grained fabric of a mudrock provides a clue as to the origin of this behaviour. Since the constituent clay minerals of a mudrock are usually encapsulated by very thin (i.e. molecular dimension) films of adsorbed water, the principal deformation mechanism is likely to be fluid-assisted interparticle shearing, possibly accompanied by particle rotation (Maltman, 1987), pressure-solutioning, and recrystallization. Small-scale cataclastic or plastic deformation of interparticle cements (e.g. calcite) and the shearing of viscous organic phases (e.g. kerogene) may be contributory
factors in some mudrocks. As is the case for many rocks, water must play a very central role in the plastic deformation of a mudrock.

In an overconsolidated mudrock, the brittle-ductile transition manifests itself in triaxial experiments performed in the laboratory as the gradual disappearance of the post-peak strain-softening response with increasing effective stress and an associated change from a highly-localized shear deformation to a more (macroscopically) homogeneous form of deformation (Horsemam et al., 1993). This would seem to be analogous to the gradual change from a dilatant to a compacting behaviour when a soil moves from the “dry side” to the “wet side” of the CSL.

If, as we suggest, significant fluid flow in a compact mudrock requires the development of dilated pathways through the material, then it seems clear that an overconsolidated mudrock on the “dry side” of critical will be more susceptible to the development of such pathways than a similar mudrock lying on the “wet side”. Furthermore, since the capacity of a clay to deform in a plastic manner decreases as its degree of overconsolidation increases, we might anticipate that pathway sealing would be more effective in normally-consolidated and lightly-overconsolidated mudrocks than in heavily-overconsolidated materials.

Relationship between critical depth and the critical state

It seems a fairly obvious extension of these ideas to suggest that there may be a relationship between the critical depth apparent in the data of Neuzil et al. (1984) and the critical state of theoretical soil mechanics. The Cretaceous shales of S. Dakota are overconsolidated. Neuzil (1993) notes that uplift and erosion have dominated the latest phase of the region’s geological history. By projecting erosional remnants it has been estimated that 360 m of overburden have been removed from the Pierre Shale in the region of one of the study sites, while geotechnical tests on borehole cores suggest 400 m of missing overburden. Stress measurements in the top 170 m of the shale show that the horizontal principal stresses are approximately lithostatic and nearly equal to the vertical stress (i.e. $K_0$ (o.c.) $\approx 1$). This provides additional evidence of overconsolidation. With reference to Figure 1, the shale has attempted to follow a path from point (c’) in the direction of (d’) on the void ratio - effective stress diagram. The development of subnormal pore pressures in the shale (Neuzil, 1993) is indicative of a state of hydraulic disequilibrium (i.e. only partially-drained conditions). Although this leads to certain complications in interpretation, as we move deeper into the formation we would expect to gradually pass from the “dry-side” towards the “wet-side”, passing through the critical state at some specific value of depth (and equivalent effective stress). The anticipated transition in material behaviour may be reflected in the hydraulic conductivity data of Figure 2.

Stress-relief fracturing

The development of fractures and fissures during stress relief is well known in overconsolidated clays and mudrocks. Such fractures develop when the magnitude of one of the principal stresses becomes substantially less than the other two. These fractures often tend
to be planar and are generally orientated so that the axis of the minor principal stress is normal to the fracture plane. They are probably associated with rupture of the fabric under extensile strain. It is important to note that extensile failure can occur even when all three of the principal stresses are compressive. The erosion and exhumation of a mudrock often leads to the development of natural sub-horizontal stress-relief fractures. Similar fractures are developed during tunnelling operations in fairly-indurated mudrocks, where they tend to be orientated sub-parallel to the tunnel walls. Because of the low stress acting normal to these discontinuities, extensile fractures are likely to act as conductive preferential pathways for the movement of groundwater and gases.

Fissures are small non-systematic joints that occur in most overconsolidated clays. The absence of shearing is thought to be indicative of brittle (extensile?) fracturing, suggesting that stress-relief due to erosion is important. A particularly important general observation is that fissure spacing tends to increase with depth (Burland et al., 1978). Groundwater movement through fissures in highly-overconsolidated clays has long been suspected.

**Intermittent or episodic flow**

The question arises as to how a fluid moves through a mudrock at a depth greater than the critical depth for pathway sealing? This is the situation of “forced-advection” alluded to in the above discussions. Our conceptual model suggests that the pressure in the source reservoir will rise until the sealing criterion is no longer met and the incipient flow pathway dilates. The flow rate along this pathway will then exhibit a highly nonlinear dependency on \( (\sigma_h - p) \), with the flow rate increasing dramatically as the source pressure continues to rise.

![Figure 6 - Schematic of the intermittent flow cycle occurring in a mudrock subject to a state of stress that favours resealing of the pathway.](image)

Eventually the source reservoir may become depleted and may depressurize. The flow to the sink will then drop off dramatically and it is possible that the pathway will reseal. Provided that the source reservoir is recharged in some way (e.g., gas generation), this cycle of quiescence followed by an active period of flow can be repeated leading to the intermittent flow
responses illustrated by Figures 6 and 7. The basic requirements for the occurrence of this intermittent or episodic flow process may be summarised as: (a) a nonlinearity and/or threshold in the flow law, (b) partial or complete resealing of the flow pathway under a suitable stress state, (c) a source reservoir capable of recharging (i.e. repressurizing) with time, and (d) sufficient energy stored in the source reservoir to support the flow during active periods. Since a compressed gas stores substantially more energy than a liquid, gas-dominated systems are likely to be much more prone to episodic flow than liquid-dominated systems.

![Diagram of source pressure, pathway aperture, and fluid flux over time](image)

**Figure 7** - Sketch of the dynamics of the intermittent flow process. The pressure in the source reservoir rises until the threshold is exceeded and the pathway dilates (point a). Flow continues until the source pressure declines and the pathway reseals (point b).

### Pockmarks in seabed sediments

Pockmarks are crater-like depressions on the seabed. They are often circular or elliptical in shape and vary in size from some 10’s to 100’s of metres across. They occur in many offshore locations throughout the world and their presence is usually indicative of underlying hydrocarbon reserves. There seems little doubt that most pockmarks are formed when thermogenic gases (largely methane), leaking upwards along discrete pathways (e.g. faults) in the underlying formations, pass through the softer and generally more deformable sediments of the seabed\(^3\) (Hovland and Judd, 1988). Although pockmarks are often scattered on the seabed, in some localities they can be arranged in a more or less linear patterns (“strings”) which probably define the strike of underlying faults and fractures.

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\(^3\) In the North Sea these are often interbedded Quaternary sands, silts and clays. In many instances the clays have been overconsolidated by a cycle of ice-loading and retreat.
The widely-accepted explanation of the formation of pockmarks is that gas pressure builds up in a shallow reservoir within the layer of soft sediments. Gas migration through this layer occurs at some critical degree of overpressuring. Migration pathways are thought to follow paths of least resistance and a single pathway may drain gas from a substantial area. Gas release is a sudden event and is often quite violent (Hovland and Judd, 1988). Plumes of sediment, some extending upwards within the water column for more than 100 m, have been observed in shallow seismic profiles (McQuillan et al., 1979).

**Figure 8** - Pockmarks are formed when thermogenic gases (largely methane), leaking upwards along discrete pathways in the underlying formations, pass through the more deformable sediments of the seabed. The gas migration mechanism is intermittent and the pathway reseals at the end of an active period.

An important feature of pockmark formation is that the gas migration mechanism is intermittent. Cycles of activity alternate with periods of quiescence. It is envisaged that, once sufficient pressure has built up and a new pathway has been established, most of the contents of a shallow gas reservoir will be emptied in a single event. During this event the gas pressure will dissipate, rapidly at first and then more slowly. The resulting reduction in pressure will increase the pressure gradient between this and deeper reservoirs, producing an upward migration of the gas and initiating another cycle of gas escape (Hovland and Judd, 1988). Although the periodicity of the process is difficult to establish, repeat surveys of pockmark locations suggests that the duration of the cycle might be around 2 to 3 years. Pockmarks can form very rapidly. In one documented case, a feature 600 m across and 30 m deep formed in 5 days. Examination of gas-bearing seabed sediments has revealed that the gas becomes trapped in cavities or vesicles, sometimes ovoid in shape, with dimensions orders of magnitude larger than normal.
pores of the sediment. Such cavities are totally enclosed by water-saturated sediment. Hovland and Judd (1988) provide a rough estimate of the methane leakage through the seabed of the North Sea, based on observations made on one continuously seeping pockmark. The pockmark measured 700 m x 450 m and produced 3000 L.hr⁻¹ of methane (at atmospheric pressure). High resolution seismic profiling suggests that this single feature drained an area of 640,000 m². The average methane flux over this area is therefore 0.04 m³.m⁻².yr⁻¹ (at atmospheric pressure) or 0.026 kg.m⁻².yr⁻¹. This is more than three orders of magnitude larger than the maximum theoretical diffusive flux of methane.

**Primary migration of oil and gas**

Hedberg (1974) provides us with a translation of a very important passage from a paper by Tissot and Pelet (1971) which states: “The displacement of an oil or gas phase from the centre of a finely-divided argillaceous matrix goes against the laws of capillarity and is in principle impossible. This barrier can however be broken in one way. The pressure within the fluids formed in the pores of the source-rock increases constantly as the products of the evolution of the kerogene are formed. If this pressure comes to exceed the mechanical resistance of the rock, microfissures will be produced which are many orders of size greater than the natural (pore) channels of the rock, and will permit the escape of an oil or gas phase, until the pressure has fallen below the threshold which allows the fissures to be filled and a new cycle commences”.

There is substantial evidence to support the fracture hypothesis of primary hydrocarbon migration (Duppenbecker, 1991; Mandl and Harkness, 1987). Microfractures containing bitumen and calcite have been observed in a number of organic-rich source-rocks (Comer and Hinch, 1987). Meissner (1978) describes oil-filled fractures in the overpressured Bakken Shale formation of the Williston Basin of N. Dakota. Mature samples of Poseidon Shale from the Lower Saxon Basin of Germany also exhibit micro and macrofractures (> 200 μm aperture) parallel to bedding (Leythaeusser et al., 1988). Meissner (1978) and du Rouchet (1981) have used the classical theories of rock failure as the basis of their analyses of microfracture development, but these theories do not explain the prevalence of sub-horizontal fractures in source-rock shales.

**Hydraulic and tectonic fractures in mudrocks**

Engelder and Oertel (1985) distinguish two types of joints that can form under abnormally high pore pressure conditions: tectonic and hydraulic. Although this distinction is somewhat artificial, it is made on the grounds that two forms of compaction can lead to high pore pressures in a sedimentary basin. Hydraulic joints form under the influence of abnormal pressures developed solely during burial and compaction. A major tectonic compression is not involved. In contrast, tectonic joints develop during orogenic events which cause lateral shortening with the volume loss producing a pressure increment beyond that due to compaction alone. This increment of fluid pressure raises the local tensile stress to the point of joint propagation. Based on indirect evidence of palaeo-overpressuring in the Devonian sediments of New York State, Engelder and Oertel have proposed a correlation between high pore pressures and the formation of cross-fold joints which are characteristic of these rocks.
**Figure 9** - Schematic plot (not to scale) of porosity versus current depth of burial showing some important trends in mudrock behaviour. Uplift, erosion and stress-relief predispose an argillaceous medium to dilatancy and "pathway flow". The realms of seabed pockmarks (= A), radioactive waste repositories (= B) and oil and gas migration (= C) are sketched.

**Implications for radioactive waste disposal**

Figure 9 summarises some of the concepts introduced in this paper and is constructed from Figure 5 by substituting depth for effective stress and porosity for void ratio. No attempt has been made to accurately scale this diagram. The zones of probable pathway sealing and of pathway dilation are distinguished by the shading. All rocks within the shaded areas are overconsolidated. Rocks lying within the upper part of the domain (marked "zone of stress-relief") are highly-overconsolidated and very prone to the development of dilated flow pathways. All rock types are capable of self-sealing given sufficient depth of burial, although the weakly-metamorphosed argillites will only seal at depths (or equivalent effective stresses and temperatures) which are quite impractical for repository construction. In general, the lower the water content of the mudrock, the greater will be the critical depth for effective pathway sealing. The critical depth for the soft clays of the seabed may be only a few metres.

The possibility of the intermittent flow of the gases generated in a repository by corrosion, radiolysis and the degradation of organic materials has been examined by Horseman and Harrington (1994) and Volckaert et al. (1995) and additional experimental research is presently underway.
Conclusions

The processes of uplift, erosion and exhumation of a mudrock, leading to overconsolidation and associated stress-relief, are evidently very important considerations when examining the propensity for fluid flow along preferential pathways in such materials. Stress-relief can produce fractures in an overconsolidated mudrock. If one of the principal stresses becomes substantially less than the other two, the mudrock will exhibit extensile straining of the fabric. If extreme, this can lead to the development of stress-relief fractures which are orientated normal to the minor principal stress. It is possible that the three-dimension networks of fissures seen in heavily-overconsolidated clays are a product of a more general form of stress-relief. During erosion, the vertical effective stress acting in a mudrock will decrease in line with the reduction of overburden thickness but the horizontal effective stress may remain high. The maximum value for the coefficient of passive earth pressure, $K_0$ (o.c.), is probably limited by shear strength. The stress of stress developed in a highly-overconsolidated mudrock at shallow depths can therefore lead to spontaneous shear failure and the formation of small reverse faults.

Beds of compact mudrock should represent very significant barriers to fluid migration. However, if the forces (fluid potential gradients) which drive flow rise to the point where they exceed the resistance of these barrier formations, then fluids will migrate by exploiting “preferential pathways”. In many cases, the incipient pathways of fluid flow are already present as faults or as interconnecting networks of smaller fractures or fissures. Under special circumstances (e.g. overpressuring), very high fluid pressures can actually create the pathways of fluid migration through mudrock formations.

Critical state soil mechanics offers a quantitative approach to the deformation, failure and pore pressure responses of a broad spectrum of argillaceous media. If significant fluid flow in a compact mudrock requires the development of dilated pathways, then it is seems clear that an overconsolidated mudrock on the “dry side” of critical will be more susceptible to the development of such pathways than a similar mudrock lying on the “wet side”. Furthermore, since the capacity of a mudrock to deform in a plastic manner decreases as its degree of overconsolidation increases, we anticipate that pathway sealing would be more effective in normally-consolidated and lightly-overconsolidated mudrocks than in heavily-overconsolidated materials. Extrapolation of the hydraulic conductivity depth-trends for the overconsolidated Cretaceous shales of S. Dakota leads to the observation that the trend for local-scale (i.e. matrix) hydraulic conductivity converges with the regional trend at a critical depth of around 2200 m. This suggests that the fractures responsible for scale-dependency would be closed at this depth. We propose a possible relationship between this critical depth and the critical state and note that a depth-related transition in material behaviour might explain the depth-trends in hydraulic conductivity observed in these materials.

The question arises as to how a fluid moves through a mudrock at depths greater than the critical depth for pathway sealing? Our simple conceptual model suggests that the fluid pressure will rise until the sealing criterion is no longer met and an incipient flow pathway dilates. The flow rate will increase dramatically if the pressure continues to rise. Eventually the source reservoir may become depleted and may depressurize. The flow will then drop off dramatically and it is possible that the pathway will reseal. Provided that the source reservoir is
recharged in some way (e.g. by gas generation), this cycle of quiescence followed by a period of flow can be repeated. Because of the very large amounts of energy stored in a compressed gas and the existence of a well-defined capillary threshold for gas entry, the episodic flow of gases is likely to be very common in geological systems.

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Generation and Dissipation of Overpressure in Sediments
Modelling and In-Situ Measurement of the Tightness of
Sedimentary Rocks

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Abstract

In many sedimentary basins, natural fluid pressures significantly greater than their hydrostatic value are observed. These overpressured zones are generally related to the presence of relatively thick impermeable and deformable clay layers. They mainly occur in the deeper part of the basins but they are occasionally recognized at relatively shallow depth (less than 1 km). This overpressure phenomenon has been widely studied in the context of the oil industry because its occurrence has drastic consequences on drilling security. Moreover, the mechanism of overpressure and the corresponding hydrodynamics are clearly related to the maturation and migration of hydrocarbons, and therefore their understanding gives fundamental clues for oil and gas exploration. In the context of waste disposals, the hydrodynamical observations of the oil industry and the sedimentary basin models which have been developed thereafter, may be of great help to i) assess the major mechanisms which affect the repositories at geological time-scale and ii) obtain significant values of the hydraulic conductivity at the scale of the massif.

Overpressure would occur if the medium has a low hydraulic conductivity. This is actually the case when impervious layers of clay are present. A wide variety of mechanisms have been proposed for generating overpressure. These mechanisms are associated with a reduction of pore volume (under gravity or tectonic loading) or/and an increase of fluid volume (associated with thermal expansion or chemical modifications). The evolution of the pressure field as a function of geological time and that of the various couplings can be described by simplified thermo-hydro-mechanical models (basin models) where the deformation is assumed to remain vertical. Using basin models, it can be shown that the reduction of pore volume by increasing effective stress - of gravity or tectonic origin- is potentially the major cause of overpressure generation. The other mechanisms act to increase the fluid volume and have generally a smaller effect. In some sedimentary basins, there is some geological evidence that chemical -mineral or organic- reactions can play some role on overpressure. Mineral transformations such as the smectite to illite reaction result in fluid production and also in pore clogging and permeability decrease (the so-called diagenetic effect). Moreover the maturation of organic matter may generate overpressure by fluid production and also relative permeability decrease when gas production is occurring. Since the reaction rate of the various chemical reactions is thermally activated, these phenomena are also likely to influence the hydrodynamics in the vicinity of nuclear waste repositories.

In sedimentary basins, the observed fact that large overpressures can be maintained over long period of time gives estimates of upper bounds for the hydraulic conductivity. This is of particular importance for argillaceous rocks, the effective hydraulic conductivity of which is very difficult to assess from laboratory measurements because of the possible fissuration of the medium. Simple analytic models are applied to the study of overpressure relaxation in sand/shale layer complex. The time constant is discussed in terms of the permeability and storativity of sand and shale materials. In the case of waste disposals, the hydraulic conductivity should also be estimated at the scale of the formation. A method of hydraulic pumping, which allows to estimate the permeability of medium, is presented. It can detect the presence of fissuration and even measure the fissuration density.
I. - Introduction

The potential safety of nuclear waste disposals in underground formations is largely dependent on the sealing of these formations. For this reason, argillaceous formations which are quite abundant in sedimentary basins offer a very interesting target. However, their transfer properties over relatively long time scales remain to be assessed.

In the past decades, much effort was devoted to the study of hydrodynamics in sedimentary basins at various scales. The rationale for these studies is mainly the understanding of the hydrocarbon maturation and migration processes and their relation to the topic of permeability of clay rocks is obvious. The comprehensive study of sedimentary basin evolution offers also a framework and a data base which is useful for predicting the large scale migration of fluids in relation with waste disposal.

In many sedimentary basins, the deep fluid pressure is largely in excess of its hydrostatic value [1]. These overpressured zones are generally related to the presence of relatively thick impermeable clay layers [2]. The occurrence and evolution of overpressure in sedimentary basins can be grossly explained by simple models taking into account the deformation of sediments and the variation of their physical properties at the time scale of the formation of the basin (i.e. Millions of years) [3] but many questions remains unanswered.

The understanding of mechanisms by which overpressures occur and evolve in sedimentary rocks is relevant to the study of wastedisposal since it characterizes a particular type of deep hydrodynamic regimes. Moreover, it can directly yield constraints on the value of hydraulic conductivity over geological time period and this value may be compared to various laboratory estimates of hydraulic properties. Besides, direct methods for estimating in situ hydraulic permeabilities in impervious rocks have also been designed and one of them, the harmonic pumping test, is of great interest for estimating relevant hydraulic properties.

In the following we first present a brief review of current knowledge concerning the occurrence and mechanism of overpressure. Then we discuss the implications of this phenomenon on the hydraulic properties of argillaceous formations and in particular the hydraulic permeability. This discussion is based on computing the time constant of overpressure relaxation in sand/shale complexes. Finally we present the method of hydraulic pumping and its possibilities for in situ measurements of the hydraulic properties in very impervious formations.

II. - Occurrence and generation of overpressure

An overpressured formation is defined as one in which fluid pressure exceeds significantly its hydrostatic value. The occurrence of overpressure in the Gulf Coast has been recognized at the beginning of the century. Since then, this phenomenon was observed in many sedimentary basins and its geographical distribution seems to be worldwide [1],[4],[5]. Figure 1 illustrates the main areas where overpressure are found. Although they are more often observed in recent sedimentary formations, overpressure are occasionally observed in other formations including paleozoic ones and in very different lithologies.

The vertical distribution of overpressure can be sketched as illustrated on figure 2. In the shallow underground, the fluid pressure is near hydrostatic and overpressure is quite weak. Below, in transition zone, the pressure begins to increase with a vertical gradient much steeper that the hydrostatic one. The underlying overpressured zone is characterized by a pressure gradient which may be hydrostatic or slightly above the hydrostatic gradient.

The mechanisms of overpressure generation have been the object of many propositions and conjectures [6]. The proposed mechanisms include: i) decrease of the pore volume by mechanical deformation under gravity loading (compaction) or tectonic compression, ii) increase of the volume of fluid by thermal dilatation, dehydration of hydrated minerals or organic diagenesis. In any case, overpressure can be generated only if the medium is sealed enough so that the overpressure producing mechanisms can compete with the overpressure relaxation due to
expulsion of fluids. The existence of low permeability formations is thus a prerequisite to the phenomenon.

The existence of a seal, i.e. an impervious formation, which prevents the overpressured fluid to escape and the excess pressure to relax, is a basic requirement for explaining the existence of overpressured zones. From lithological point of view, pressure seals may be either initially impervious rocks like clays or salt or other types of porous rock which have lost their permeability by diagenetic process. Following the work of Bradley [7], Pawley [8] and Hunt [9] on the existence of sharply limited pressure compartments, the existence of diagenetic pressure seals has been largely debated [10]. However there is no doubt that argillaceous formations may play an important role in the occurrence of overpressure because of their impervious nature and because of their mechanical ductility.

Since the overpressure generation and evolution is a progressive phenomenon, it can be studied by modelling the various physico-chemical phenomena occurring during the evolution of the sedimentary basin. The interest of basin modelling is that it can take into account the various coupled mechanisms and allows to discuss their relative importance. The equation describing the expulsion of fluids from geopressured sediments can be written [12],[13]:

\[
\frac{d\phi}{d\sigma} \frac{d\phi}{d\sigma} \frac{dP}{dt} = \frac{\partial}{\partial z} \left( k \left( \frac{\partial P}{\partial z} - \rho g \right) \right) + \frac{d\phi}{d\sigma} \frac{d\sigma}{dt} + \alpha \phi \frac{\partial T}{\partial t} + q \quad (1)
\]

\(d/dt\) stands for the derivative following the matrix movement. \(P\) is the fluid pressure as a function of time \(t\) and depth \(z\), \(\phi\) the porosity, \(\Sigma\) the vertical stress, \(T\) the temperature, \(g\) the acceleration of gravity and \(q\) a source term accounting for volumic fluid production. \(\beta\) is the compressibility of the fluid, \(\rho\) its specific mass, \(\alpha\) its thermal dilatation coefficient, and \(\mu\) its viscosity. The medium is characterized by its hydraulic permeability \(k\) and by the constitutive relation between the porosity \(\phi\) and the vertical component of the effective stress \((\sigma = \Sigma - p)\) from which the compressibility \(d\phi/d\sigma\) is deduced. From a mechanical point of view, the underlying assumption is that the medium is poro-plastic or elasto-poro-plastic. The progressive burial of rocks during sedimentation is accounted for by the term \(d\Sigma/dt\) (rate of vertical stress increase) as well as by the term \(dT/dt\) (rate of temperature increase). The permeability \(k\) is generally assumed to be related to the porosity by a power law:

\[k = a\phi^n\]  

(2)

Overpressure is generated in the system at the condition that fluid pressure builds up at a rate faster than it vanishes under diffusion. Therefore thick impervious clay sequences and large sedimentation rate appears as favorable conditions for overpressure generation. In such circumstances the main mechanism by which overpressure is generated is the compaction of deformable porous sediments (i.e. clay rocks) under the effect of burial load. The fluid pressure evolution in a sedimenting thick clay layer is presented on figure 3 from Bredehoef and Hanshaw [5]. This result uses simplifying assumptions (constant permeability, non deformable matrix rock...) and an analytical solution formerly obtained by Gibson. It is clear from figure 3 that, when the hydraulic conductivity is small enough, the calculated fluid pressure evolution departs from its hydrostatic trend and reaches quite large values at the bottom of the profile. However the fluid pressure remains everywhere smaller than the lithostatic value. For this problem, Audet and Mac Connell [14] and Wangen [15] defined a useful dimensionless number which rules the occurrence of overpressure:

\[\lambda = K / V_0 (\rho_s / \rho - 1)\]  

(3)

where \(\rho_s\) is the solid grain density, \(K = (k\rho g / \mu)\), the hydraulic conductivity at some reference level and \(V_0\), the sedimentation rate (which is proportional to \(d\Sigma/dt\) in eq.(1)). For \(\lambda >> 1\), the medium is so permeable or/and the loading rate so low that overpressure does not build up. With decreasing \(\lambda\), the fluid pressure increases and, for small \(\lambda\) value, reaches asymptotically the lithostatic pressure.

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Using numerical solutions of eq. (1), it is possible to simulate the relative importance of other processes that burial and compaction on the generation of overpressure. Figure 4 from Luo [6] allows to compare the influence of various overpressuring mechanisms with that of compaction. The effect of water dilatation under temperature increase (aquathermal effect) as well as that of bound water release during the diagenesis of smectitic clay minerals appear completely negligible as far as only they volumetric effect is concerned [13]. On the contrary, the influence of lateral tectonic stress may be important as well as that of organic diagenesis in the case where kerogen and oil are cracked into gas [16]. This latter mechanism is due to the simultaneous effect of fluid specific volume increase and of relative permeability decrease of the two phase fluid [16].

This type of numerical model which can be used either in 1-D or in 2 or even 3-D [1],[12],[17],[18],[19], accounts quite satisfactorily for the main features of observed overpressure or inferred hydrodynamics. A classical example is the Gulf of Mexico where fluid overpressures are found to be the result of rapid subsidence and sedimentation during the past 2My [20]. However it must be realised that the proposed models are extremely dependent on the assumed value of permeability as illustrated in figure 3.

III. - Constraints on the hydraulic conductivity from overpressure data

The occurrence of overpressure in a given formation depends mainly on the sealing efficiency of this formation and/or of neighbouring ones. The question of what rock permeability is required for maintaining anomalous pressure has been the object of some recent polemical papers [21],[22],[23]. Using analytical solution to the problem of pressure relaxation, Deming [21] estimated that the permeability value necessary to maintain overpressure over a period span of about 1 My is in the range 10^{-22} - 10^{-23} m^2, i.e. lower than most laboratory measurements of shale permeability. However, He and Corrigan [22] pointed out some inaccuracies in the model developed by Deming [21]. In the following, the same type of arguments is developed, but we present 1-D analytical models of shale/sand complexes which are realistic representations of many geological situations.

The problem addressed is illustrated in figure 5 and may be summarized as follows: consider an impervious shale layer which isolates a lower permeable sand layer from the upper formations which are also permeable. Assume that, initially at time t = 0, the lower formation is overpressured with constant excess pressure. In the insulating shale layer, the initial overpressure is assumed to vary linearly with depth. The upper boundary condition is of zero overpressure at the top of the shale layer and of zero flux at the bottom of the system.

In the following, both layers are assumed homogeneous and the various parameters (compressibility, permeability, viscosity ...) are constant. No specific volume variation, no source effect and no loading effect are considered and the geometric deformation of the medium is neglected. Following hydrogeologic tradition [24], we introduce the hydraulic head $H$, the specific storage $S$ and the hydraulic conductivity $K$ defined as:

$$H = \frac{P}{\rho g} - z \quad (4)$$

$$S = \left( \phi \beta + \frac{d\Phi}{d\sigma} \right) \frac{\rho g}{1 - \phi} \quad (5)$$

$$K = \frac{k \rho g}{\mu} \quad (6)$$

The equation (1) then reduces to the classical diffusion equation:

$$S \frac{\partial H}{\partial t} = K \frac{\partial^2 H}{\partial z^2} \quad (7)$$

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For the geometry illustrated in figure 5 we call \( S' \) and \( K' \) the storage coefficient and the hydraulic conductivity of the shale layer, \( L' \) its thickness. In the underlying sand layer of thickness \( L \) and storage coefficient \( S \), the hydraulic conductivity is assumed much larger than in the shale layer so that the vertical gradient of \( H \) is very small. If we integrate eq.(7) on the thickness of the sand layer, we obtain:

\[
S' \int_0^L \frac{\partial H}{\partial z} \, dz \approx SL \frac{\partial H}{\partial t} = K \frac{\partial H}{\partial z} \bigg|_{z=L} - K' \frac{\partial H}{\partial z} \bigg|_{z=0} \tag{8}
\]

Now the condition of no flux at the base of the sand imposes: \( 0 = K \frac{\partial H}{\partial z} \bigg|_{z=L} \) whereas the continuity of fluxes between the clay layer and the sand one gives: \( -K \frac{\partial H}{\partial z} \bigg|_{z=0} = -K' \frac{\partial H}{\partial z} \bigg|_{z=0} \). Thus, the equation (7) for the sand layer may be replaced by [25] :

\[
S \frac{\partial H}{\partial t} = -\frac{K'}{l} \frac{\partial H}{\partial z} \bigg|_{z=0} \tag{9}
\]

The solution to this initial value problem, with initial conditions illustrated in figure 5, may be obtained by Laplace transform. Defining the Laplace transform \( \bar{H} = \int_0^\infty He^{-pt} \, dt \), in the sand layer one obtains, with \( s' = \sqrt{\frac{S'}{K'}} \):

\[
\bar{H} = H_0 \frac{S - \frac{K'}{pl''}(1-s'l' \coth(s'l'))}{S + \frac{K's'l'}{pl''} \coth(s'l')} \tag{10}
\]

The behaviour of this expression may be simplified for large \( t \) value, considering the limit \( p \to 0 \). Since \( x \coth x \to 1 + x^2/3 \) for small \( x \) value, we obtain:

\[
\bar{H}(p \to 0) \approx H_0 \frac{1 + \frac{S'l''}{3Sl}}{\left(1 + \frac{S'l'}{3Sl}\right)p + \frac{K'}{3Sl}} \tag{11}
\]

The inverse Laplace transform gives the asymptotic behaviour:

\[
H(t \to \infty) \approx H_0 e^{-\tau/t} \tag{12}
\]

\[
t = \frac{S'l''}{K'} \left(1 + \frac{S'l'}{3Sl}\right) = \frac{l^2S'}{3K'} \left(1 + \frac{3Sl}{S'l'}\right) \tag{13}
\]

This functional dependence on the geometric and hydraulic parameters of both layers presents interesting properties. When \( S'l' \ll Sl \) (i.e. when the sand layer is thin or with small specific storage), the time constant is the classical characteristic diffusion time of the insulating layer. However, when \( Sl \) becomes significant, it increases the value of the time constant \( t \) by a factor \( (1 + 3Sl / S'l') \) which can be significantly greater than \( 1 \).

Coming back to the problem of relaxation, we now discuss the value of the hydraulic parameters which are required by the maintenance of overpressure for a given time span, say \( t = t_1 \) My through an insulating clay layer of thickness \( t' = 100 \) m. For a given value of \( Sl/S'l' \), the constrained quantity is the value of \( K'/S' \), i.e. the hydraulic diffusivity. If \( Sl / S'l' = 0 \) then the required hydraulic diffusivity is about \( 10^{-10} \) m²s⁻¹ i.e. a very small value.
Values of the specific storage related to this problem may be estimated from the observed compaction of sediments as a function of depth in geological formations where hydrostatic equilibrium prevails. Assume for sake of simplicity an exponential decrease of porosity v.s. depth (the so-called Athy’s law [26]):

$$\phi = \phi_0 e^{-c_0}$$

(14)

$\phi_0$ is the surface porosity and $c_0$ rules decrease of $\phi$. This depth dependence reflects a dependence with the vertical effective stress defined as $\sigma = (\rho_b - \rho)gz$, $\rho_b$ being the bulk density of sediment plus water, Eq. (14) can be written:

$$\phi(\sigma) = \phi_0 e^{-\frac{c_0}{(\rho_b - \rho)}}$$

(15)

Neglecting in eq. (5) the compressibility of water with respect to that of the bulk rock, the storage coefficient $S'$ becomes:

$$S' = \frac{1}{1 - \phi} \frac{d\phi}{d\sigma} \rho g = \frac{\phi}{1 - \phi} \frac{c \rho}{\rho_b - \rho}$$

(16)

For deep shale formations, typical values are $c = 10^{-3}$ $m^{-1}$ [27],[28], $\rho_b = 2.3103$ $kg m^{-3}$, $\phi = 0.2$ and $K' \approx 2.10^{-14} m s^{-1}$. An hydraulic diffusivity of $10^{-10}$ $m^2 s^{-1}$ is achieved for a conductivity $K' \approx 2.10^{-14} m s^{-1}$. With a water viscosity of $0.4 \times 10^{-3}$ Pa s, such a conductivity corresponds to a permeability $k = 0.8 \times 10^{-21} m^2$ which is an extremely low value (less than a nanoDarcy), at the lower limit of laboratory measurements [21].

However, when the storativity of the bottom layer (Fig 5) is taken into account ($S' \approx S' l'$ the requirement of low diffusivity and low permeability in the insulating layer becomes less severe. Assume a bottom sand layer with specific storage $S = 0.3 \times 10^{-3}$ $m^{-1}$ (it is usually smaller than for clay) and thickness $l = 1000$ m. Then a permeability $k = 0.8 \times 10^{-20} m^2$ is obtained which is in better agreement with most laboratory values [29].

The assumptions used for the relaxation of overpressure in a single sand/shale complex (Figure 5) are very restrictive and natural situations are much more complex. A useful model is that of a sandwich composed of n sand/shale alternating layers as shown in figure 6. The corresponding equations are (with obvious notations).

$$S_n \frac{\partial H_n}{\partial t} = K_n \frac{\partial H_n}{\partial z} \bigg|_{z_n} - K_{n-1} \frac{\partial H_{n-1}}{\partial z} \bigg|_{z_{n-1} + b_{n-1}}$$

(17)

$$S_n \frac{\partial^2 H_n}{\partial t^2} = K_n \frac{\partial^2 H_n}{\partial z^2}$$

(18)

in the sand layer and in the shale layer respectively. The general solution of the initial value problem, which starts from initial constant overpressure gradient in the different shale layers, can also be obtained by Laplace transform of eqs. (17-18). A relation between $\bar{H}_{n-1}, \bar{H}_n, \bar{H}_{n+1}$, the overpressures in the sand layers of index n-1, n, n+1, is obtained. This relation can be written matricially as:

$$(p \bar{\alpha} + \bar{\beta}) \bar{H} = \bar{\alpha} \bar{H}_0$$

(19)

where $\bar{H}$ is the vector $(\bar{H}_1, ... , \bar{H}_n)^T$, $\bar{\alpha}, \bar{\beta}$, two symmetric, positive definite tridiagonal matrices.

Using the eigenvector decomposition of the matrix $\bar{\alpha}^{-1} \bar{\beta}$:

$$U \: \bar{\alpha}^{-1} \bar{\beta} U = \Lambda$$

(20)

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where $\Lambda$ is a positive diagonal matrix of eigenvalues ($\lambda_i$) and $\bar{U}$ the matrix of eigenvectors. If $\bar{Q}$ and $\bar{Q}_0$ are defined as:

$$\bar{Q} = \bar{U}^{-1} \bar{H} \quad \text{and} \quad \bar{Q}_0 = \bar{U}^{-1} \bar{H}_0$$

then, one obtains for the component of $\bar{Q}$:

$$\bar{Q}_i = \frac{Q_{0,i}}{\lambda_i + p}$$

which can be Laplace- inverted in the time domain:

$$Q_i(t) = Q_{0,i} e^{-\lambda_i t}$$

with $\tau_i = 1/\lambda_i$. Using the diagonal matrix $\bar{E}$ whose $i$th element is $e^{-t/\lambda_i}$, this can be written:

$$\bar{Q} = \bar{E} \cdot \bar{Q}_0$$

For large $t$ value, the mode corresponding to the smallest eigenvalue (largest time constant $t_m$) becomes rapidly dominant. Numerical computations of $t_m$ have been performed in the case where all layers have similar geometries and properties (i.e. $S'_i = S'$, $l'_i = l'$, $S_i = S$, $l_i = l$, $K'_i = K'$). It was found that, for large number of shale/sand layers, the largest time constant $t_m$ can be expressed by:

$$t_m = \frac{n^2 ll' S}{2.5 K'} \left( 1 + \frac{S' l'}{S l} \right)$$

This result is a generalization of the previous one for a single sand/shale layer complex. It can be interpreted by saying that a suite of $n$ shale/sand complex of thicknesses $l$ and $l'$ behaves as a single one of thickness $n l$ and $n l'$.

IV- In situ measurements of hydraulic conductivities by the harmonic pumping test

The direct in situ investigation of hydraulic conductivities was primarily based on permanent flow tests such as the Lugeon test. More information was obtained by pulse tests, essentially used in oil investigation. More recently the idea of harmonic pumping test was proposed to cover applications not accessible to the previous techniques.

Historically, the harmonic pumping test was imagined for investigating the extension and the aperture of hydraulic fractures created in granite at large depth in hot dry rock applications. Of course such an information cannot be given by a permanent hydraulic test. In the same manner, a pulse test is not adapted to the investigation of limited fractures because, under a sudden pressure drop, no valuable measurement is possible.

The principle of the harmonic pumping test consists in creating in a cavity of the medium to be recognized a sinusoidal quantity of flow $q$ versus time $t$, at frequency $f$ [30]. Due to the fluid compressibility, this cavity behaves as a fluid resonator where the pressure variation $p$ depends on time $t$ according to a sinusoidal law assuming the flow to be laminar. In the frequency space, this test leads to a transfer function $P(f)/Q(f)$ between the Fourier transforms of the input signal $q(t)$ and the output signal $p(t)$.

Starting from this historical case study, the harmonic pumping test has been enlarged and tools have been adapted to a number of different types of porous and/or fractured media, at different scales from the laboratory sample up to in situ formations, with a large range of hydraulic conductivities.

The identification procedure of hydraulic conductivities is based on the following steps:
(i) An in situ harmonic test is realized by creating between two packers in a well a sinusoidal flow $q(t)$ and recording the resulting sinusoidal variation $p(t)$ of the pressure in the
same cavity. The complex ratio $P(t)/Q(t)$ of the Fourier transforms of the two experimental signals $p(t)$ and $q(t)$ gives the experimental hydraulic spectral signature $S(f)$ of the medium and the cavity.

(ii) A numerical simulation of this test is carried out taking into account the type of the medium encountered and the boundary conditions. The movement of the fluid within the fractures depends on inertia, compressibility and viscosity forces. As the sinusoidal variations $p(t)$ of the pressure are generally negligible as compared to the static pressure $p_0(t)$, the harmonic pumping test is not sensitive to a possible deformation of the bulk. This modelling leads to a set of transfer functions $F(f,a_i)$ depending on the frequency $f$ and the medium parameters $a_i$.

(iii) Finally comparing the experimental spectral signature $S(f)$ with the set of theoretical transfer functions $F(f,a_i)$ leads, by a matching procedure, to an estimation of the hydraulic parameters to be recognized. This estimation relies on both modulus and phase charts.

The theoretical transfer function of a unique and closed fracture can be computed neglecting viscosity forces as compared to inertia and compressibility forces. The modulus of this transfer function exhibits a set of resonance frequencies, which depends on the extension of the fracture. Between these resonance frequencies, the values of the modulus depend on the equivalent hydraulic aperture of the fracture. The phase spectrum is reduced to a phase difference of $\pm 90^\circ$ [31],[32]. An example of such a transfer function of a fracture perpendicular to the well is shown in figure 7 (after Crosnier [33] p. 49). The transfer function is not sensitive to the inclination of the fracture on the well axis up to angles of $80^\circ$.

The theoretical transfer function of open fractures can be computed neglecting compressibility forces as compared to inertia and viscosity forces. This transfer function depends on the number and the aperture of the fractures. In a deterministic approach, different sets of fractures are distinguished. For instance, in the case of two families of parallel fractures, the number and the aperture of the fractures of both families can be obtained in a unique manner by the analysis of the transfer function. The experimental verification has been obtained on a laboratory equipment as shown in figure 8 (after Fras, Pons et Jouanna,[30] fig.5). A good agreement with theory is obtained at low frequencies as shown in figure 9 (after Fras, Pons et Jouanna, [34], fig.7). At higher frequencies, some discrepancy can be introduced by turbulence due to the fact that the quantity of flow is proportional to the frequency. Such non linear phenomena are easily avoided by adjusting the volume of the piston to the frequency range.

More recently a stochastic approach was proposed for interpreting such tests. Analysis of the transfer function deals to the moments of any order of the aperture distribution of the fractures [35][36]. The mean and the standard deviation of the aperture distribution can be rapidly obtained by interpreting only the transfer functions behaviour at low and high frequencies.

An example of the determination of hydraulic parameters of a calcareous Valanginian formation is given in Crosnier [33]. After recording raw data, the type of model to be used for simulating the in situ rock is determined. Then the chart of theoretical transfer function is drawn and the hydraulic parameters are obtained by matching. In the described example, the harmonic pumping test was realized at a depth of 8.6m to 10.0m, using a range of frequencies from 0.17Hz to 13 Hz. The data analysis leads to an open type of a unique fracture which is active on a sector of $35^\circ$ with an equivalent hydraulic aperture of 0.6mm.

V.- Conclusion

Fluid overpressure is a common hydrodynamic sedimentary basins. Its occurrence mainly stems from the increasing gravity or tectonic loading under burial and from the resulting compaction of shale layers. Other causes such as diagenesis may also play some role. In any case, the generation and the maintaining of overpressures over long geological periods (1My) imply that the medium is very tight. The required permeability of a 100 m thick shale layer which would insulate the deep hydraulic system is no more than $10^{-20}$ to $10^{-21}$ m$^2$. 

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Such low values of permeability raise the important question of fracturation which would unavoidably affect the overpressured zones when the mean effective stress decreases below some threshold. Harmonic pumping tests provide a unique way determining in-situ hydraulic conductivity and to detect potential fissures.

References

Figure 1: Hatched areas represent basins where overpressure have been encountered.

Figure 2: Sketch of the pressure profile and corresponding hydraulic head in a zone where overpressure is occurring.
Figure 3: Some pressure plots calculated by Bredehoef and Hanshaw for conditions of the Gulf Coast with various assumptions on the hydraulic conductivity (kpg/μ).

Figure 4: Influence of various mechanisms on pressure evolution during burial. The reference case corresponding to compaction acting alone is plotted as a continuous line. The effect of compaction plus another mechanism (thermal, smectite dehydration, organic matter degradation or tectonic stress) is illustrated by a dotted line.
Figure 5: Illustration of the relaxation model for a sand-shale complex. A shale layer of thickness $l'$, conductivity $K'$, storage coefficient $S'$ is overlying a sand layer with thickness $l$ and storage coefficient $S$. The initial head is $H_0$ in the sand layer and linear in the shale.

Figure 6: Illustration of the relaxation computation for a sandwich composed of several sand/shale complexes.
Figure 7: Some transfer functions of circular closed fractures. Well radius: 0.165 m. Fracture extension: 100 m. First resonance: 8.4 Hz. Fracture aperture: $2 \times 10^{-4}$ m; $10^{-3}$ m and $2 \times 10^{-3}$ m.

Figure 8: Laboratory equipment for an harmonic pumping test. Case of one or two open fractures.
Figure 9: Comparison between experience and theory (modulus of the transfer function). Case of one or two 2 mm-thick open fractures.
Recognition of the Geochemical Effects of Expulsion of Petroleum from Source Rocks

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Geologisches Institut der Universität zu Köln

Extended Abstract

Understanding petroleum migration processes has for a long time remained the single most puzzling problem of petroleum geology and geochemistry. Over the past decade considerable progress has been made, however, towards a better understanding of the mechanisms and efficiencies of flow of petroleum fluids through the pore system of source rocks of siliciclastic and of carbonate lithologies. Not the least was this progress achieved by studies of actively expelling petroleum source rocks with modern analytical methods of petroleum geochemistry. In this presentation the results of many years of geochemical studies carried out by the author’s former group at the Research Center Jülich (KFA) will be reviewed. This work includes case history studies of the geochemical effects of petroleum expulsion from mature source rocks as well as experimental studies to simulate petroleum migration under controlled laboratory conditions. Main emphasis of this review is on a discussion of concepts and methods on how to recognize and how to quantify the effects of petroleum flow in fine-grained argillaceous rocks.

Rich source rocks like the Upper Jurassic Kimmeridge Clay Formation in the North Sea area expell petroleum as a separate phase fluid. Leaner quality source rocks bearing type III kerogens expell petroleum in gaseous solution. Expulsion efficiencies vary widely and evolve with maturity progress, i.e. are related to the degree of kerogen conversion. In a detailed study of extended series of core samples from oil-prone source rocks of the Kimmeridge Clay Fm. interbedded with reservoir sandstones, the geochemical migration effects, schematically summarized in Table 1, have been observed and interpreted in terms of driving forces for fluid flow processes. The basic concept applied in this study was based on the idea that the geochemical effects of petroleum expulsion should best be recognized by comparisons of samples which have experienced different drainage conditions and hence are likely to have experienced variable migration efficiencies under natural geologic conditions. Examples of this kind were found to be high-resolution suites of samples from the center of thick source rock intervals towards their nearest contact with the over- or underlying reservoir rocks, as well as samples of thin source rock layers (10 – 30 cm) interbedded in reservoir sands. If data comparisons of such sample series are made in a quantitative way (using measured concentrations of bulk hydrocarbon fractions or individual molecular species) mass balance calculations can be carried out leading to determinations of relative expulsion efficiencies.

The molecular composition of the residual petroleum remaining in the source rock pore system after expulsion was recognized as a key factor for a better understanding of the conditions of flow of petroleum fluids. Especially, the type of migration mechanism can be identified in this way. An
example illustrating this approach is shown in Fig. 1. The molecular distribution of the saturated hydrocarbons extracted from two source rock samples, which had originally generated hydrocarbons of identical composition, reveals pronounced differences. The sample 127.0 m taken from the center portion of a thick source rock interval shows a broad bimodal distribution of the n-alkanes, which are also more abundant than the isoprenoid hydrocarbons pristane and phytane. Sample 62.5 m is from a source rock of the same organic matter–type, but occurring as a 5 cm thin interbed within reservoir sands. Its unimodal, heavy–end biased n-alkane distribution with a pronounced predominance of pristane is a result of preferential expulsion of lower molecular–weight hydrocarbons from this shale interval (shaded area in Fig. 1). A mass balance of hydrocarbon concentrations for these two samples reveals molecular fractionation effects which are indicative of transport of these hydrocarbons in gaseous solution.

In the second part of this presentation observations and conclusions about the role of micro–fractures in primary migration processes are reviewed. Case histories studied in this context include the Toarcian–age Posidonia Shale source rocks from NW–Germany as well as a Triassic–age source rock from Italy. It is shown under which circumstances development of pore fluid overpressures can lead to micro–fracturing of the rock matrix. The geochemical effects of the flow of petroleum fluids from the shale matrix into fractures have been recognized on the basis the recognition of local gradients of concentration and composition of selected hydrocarbon species.

Table 1: Summary of observations regarding geochemical migration effects for Kimmeridge Clay Fm. source rocks. Brae Field Area. British North Sea.

<table>
<thead>
<tr>
<th>Observed Effects</th>
<th>Interpreted mechanisms</th>
</tr>
</thead>
<tbody>
<tr>
<td>–Intervals of increasing depletion towards shale/sandstone–contacts (“edge effects”)</td>
<td>Capillary forces (5 – 8 m wide edges)</td>
</tr>
<tr>
<td>–Locally high depletion (thin interbedded shales)</td>
<td></td>
</tr>
<tr>
<td>–Uniform expulsion efficiencies on molecular level</td>
<td>Bulk oil expulsion as separate phase flow</td>
</tr>
<tr>
<td>–for members of homologous series (n-alkanes)</td>
<td></td>
</tr>
<tr>
<td>–among isomers (e.g. methylphenanthrenes)</td>
<td></td>
</tr>
<tr>
<td>–Non–uniform expulsion efficiencies on gross compositional basis, i.e. Saturated</td>
<td>Fractionation processes (of unknown character)</td>
</tr>
<tr>
<td>HCs &gt;&gt; NSO</td>
<td></td>
</tr>
<tr>
<td>–Local and selective enrichments with (shales with bulk oil-depletion):</td>
<td>“Back”–Diffusion from reservoir.</td>
</tr>
<tr>
<td>–light hydrocarbons (C1 – C8)</td>
<td>Imbition of water from reservoir with dissolved HCs/adsorption</td>
</tr>
<tr>
<td>–benzothiophenes</td>
<td></td>
</tr>
</tbody>
</table>

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Literature included in this review


Figure 1. Capillary gas chromatograms indicating composition of saturated C<sub>15-30</sub> hydrocarbons of two shale samples from the Paleocene Firkanten Fm. from Svalbard, Norway. Basic geochemical data are indicated. Prominent peaks are n-alkanes of indicated carbon number range. Since hydrocarbon generation conditions were the same for both source rock samples, the compositional difference reflects preferential expulsion of lower molecular-weight hydrocarbons from sample 62.5 m.
Fault Zones as Barriers to, or Conduits for, Fluid Flow in Argillaceous Formations: A Microstructural and Petrophysical Perspective.

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Abstract

Patterns of flow in sedimentary basins are often controlled by the geometry, size population and spatial distribution of faults and fractures that link or compartmentalize permeable horizons. The microstructural properties of individual faults dictate whether they act as conduits, or barriers to convective, diffusive and multiphase fluid transport. To improve quantitative predictions of the hydrogeological impact of faults, it is necessary to understand the relationship between the mechanics of rock deformation and the evolution of petrophysical properties. Fault rocks from North Sea reservoir intervals have been studied in detail using this approach. Since a wide range of fault rocks was analysed, many of the findings and techniques can be applied to lower permeability environments.

During fault slip, the microstructure of intact rock is changed by mechanical and chemical processes that together constitute the deformation mechanisms through which the rock volume around and within the fault is strained. Following a faulting event, the rock may be modified by stress relaxation and the passage of fluids through or along the fault. The physical deformation mechanisms are largely controlled by the effective stress mobilised at grain contacts: grain rolling and sliding giving way to cataclasis as stress increases. The chemically-assisted deformation mechanisms (diffusive mass transfer processes) require time for material transport to occur. Thus they are mainly manifest during stress relaxation, when pressure solution features commonly overprint cataclastic deformation. Deformation mechanisms in sediments are strongly influenced by temperature, fluid activity and the presence of phyllosilicate minerals. Clays reduce interparticle friction and depending on their type, quantity and distribution can either hinder or promote diffusive mass transport.

Deformation mechanisms all act to reduce porosity and permeability of fault rocks with respect to their precursor lithology. This effect is enhanced by post-deformational cementation, which is often concentrated in the vicinity of fault zones by steepened thermal, geochemical or fluid-pressure gradients. The faulted rock volume is also altered in its mechanical properties. Subsequent stress increases may be relieved by different deformation mechanisms, including brittle fracturing that may breach the integrity of the sealed fault zone. In this way, a fault zone that initially seals may become a conduit for sub-vertical or cross-formational fluid migration. While permeability is generally low in argillaceous formations, it may still vary by several orders of magnitude between horizons due to changes in clay content. Even thin layers of high permeability may act as important flow pathways,
and the potential for fault zones to seal or inter-link high-permeability domains must be taken into account when assessing the suitability of a particular formation for long-term waste disposal.

Introduction

Properties of Argillaceous Formations

Traditionally, the hydrogeological properties of predominantly argillaceous rock formations have received relatively little attention compared with the highly porous or permeable rocks that may serve as economically important aquifers or reservoirs. Clays and shales were generally considered to be seals or aquitards, that in comparison had negligible permeability. It is now widely appreciated that clays and shales have low but measurable matrix permeability, and that this can vary over many orders of magnitude; typically $10^{-22}$ to $10^{-15}$ m$^2$ [1]. It is this property—low permeability—that makes argillaceous deposits attractive as potential sites for surface and sub-surface waste disposal. We focus on permeability in this paper, but in very low permeability environments, much of the transport will be dominated by diffusion through the fluid phase, which is a function of porosity and tortuosity.

The controls on the permeability of argillaceous formations are very complex. Firstly, the textural and mineralogical properties of the original sediment dictate what we can call the "base-level" character of its transport properties [2,3]. Increased clay content, and among the clay minerals, increases in the proportion of mixed-layer and expandable clays both decrease the base-level permeability. These original properties then evolve over geological time through the action of mechanical and chemical processes. Mechanically speaking, fine-grained sediments, are relatively compressible elastoplastic materials [4,5] and so their porosity and permeability depend strongly both on in situ effective stresses and on previous stress history. Generally permeability is found to decrease in a log-linear way with decreasing porosity, and so the decrease in permeability with burial depth is greater in more compressible, clay-dominated sediments [6]. The geochemical evolution of argillaceous sediments can be of equal importance in determining their transport properties, as they may undergo a spectrum of early diageneric changes [7], and on deeper burial their mineralogy can be altered completely by a succession of dehydration reactions [8].

Heterogeneity and Scale Dependence of Transport Properties

As a result of the complex interactions of the processes described above, significant variations in transport properties are expected both within and between lithological units of a given formation. Textural and mineralogical properties may change on the scale of laminae to beds (mm to cm), while lithofacies change vertically and laterally on the scale of metres to kilometres. What may be subtle changes in pore structure may be reflected in significant changes in the transport properties of the rocks [9]. Faults and fractures can segment and link lithological zones with contrasting hydrogeological properties. The result is that a geological formation, which may appear as a single colour on a geological map or section, is, hydrogeologically speaking, a heterogeneous and anisotropic system of poroperm units linked and separated by conduits and barriers.

While chemical and mechanical changes to material properties can be examined in laboratory scale tests, the effects of spatial variability can only be appreciated using a sufficiently detailed flow model. Accordingly, when extrapolating permeability measurements beyond the laboratory scale or the scale of local field tests, care must be taken to chose an upscaling strategy that will give realistic (or at least conservative) results in model simulations. Since the variation in permeability is manifest on a hierarchy of scales, it is expected that over some range of observation, apparent permeability will increase progressively to a level that represents the overall transmissivity of the formation [10]. A qualitatively similar variation is to be expected in the other transport properties (diffusivity, capillary entry pressure and multi-phase conductivity).
Fault Seals: Overview and History

Faults commonly act as barriers to the flow in hydrocarbon reservoirs (described here), and can also act as conduits for the migration of hydrocarbons from source areas into the reservoirs (described below). Sealing faults can pose a serious problem for exploitation if they compartmentalize reservoirs that otherwise have good petrophysical properties. For these reasons, a considerable amount of effort has been directed to the understanding of fault sealing in reservoir settings, and this work has implications for more general problems of subsurface fluid flow.

After it became apparent that faults often produced economically important hydrocarbon traps, the understanding of fault sealing has progressed through a number of recognizable stages. Initially it was thought that faults led to sealing through the juxtaposition of a fine grained sedimentary unit—generically a “shale”—against a reservoir sand interval so that it produced a lateral barrier to hydrocarbon migration. Later it was recognised that shale or clay, being ductile, could be dragged into zone of slip so that the fault plane itself produced a “shale-smear” seal [11]. In these two instances the fine grained sediments act as a seal because of the low permeability and high capillary entry pressure to hydrocarbons inherent in the original shale lithology.

More recently it has been recognised that reservoirs can become compartmentalized by faults even when their displacement is so small that there are only juxtapositions of sand against sand, with no possibility of a shale smear occurring. Microstructural analysis of samples from these faulted sand intervals revealed that the seals were produced by thin cataclastic shear zones known as granulation seams that have significantly reduced porosity and permeability and markedly increased capillary entry pressure (see below). The recognition of cataclasis as an agent in reservoir compartmentalization led to a paradigm shift in fault seal analysis towards a greater appreciation of the mechanical processes in fault zones and their subsequent microstructural evolution.

Deformation Mechanisms in Sediments Undergoing Lithification

The grain-scale processes by which strain is accommodated in sediments, rocks and minerals are known as deformation mechanisms [12,13], (Fig. 1). As a sediment is buried and subjected to different regimes of temperature, pore fluid pressure and differential stress, strain is accommodated by a succession of mechanisms, that act singly or in tandem, to produce deformed rocks or tectonites. The physical and transport properties of the sediments are changed by the combined effects of deformation and diagenesis, and since many tectonic processes such as faulting involve hydro-mechanical coupling, these changes may feed back into the geological system in a complex way [14,15].

Independent Particulate Flow and Soft Sediment Deformation

Recently deposited sediments have little cohesion and low frictional strength. Modest shear stresses are accommodated as particles are reoriented and slide past one another in a process known as independent particulate flow. Commonly, significant volume changes may occur as particles move closer together during shear-induced compaction, or move apart from one another, resulting in dilation. The mechanical relationships between these volume changes, normal stresses and shear stresses in un lithified sediments are the realm of soil mechanics [4,5]. Geologically speaking, this regime is termed wet-sediment or soft-sediment deformation [16]; the main characteristic being that strain is accommodated while the individual grains remain intact. While deformation of this type may be distributed throughout the material, and truly ductile in the sense that there is no sudden loss of strength, geometrically and mechanically brittle processes of faulting and fracturing can also occur in the soft sediment regime.
Brittle Failure and Cataclasis

When confining stresses reach a certain level, it is no longer possible for strain to be accommodated in deforming sediments wholly by the non-destructive sliding and rolling of the constituent particles. Interparticle stresses become such that at point contacts brittle fracture is initiated, resulting in grain breakage. This leads to a reduction in the grain-size of the affected sediment, and this is enhanced by abrasion and wear within slip zones. It is common for slip to be concentrated in narrow zones (cataclastic shear zones) which become the locus of strongly reduced grain size. In these zones, high interparticle stresses have caused grain breakage, and with abrasion during slip, the particle size and porosity are reduced markedly, so producing a zone of low permeability and high capillary entry pressure [17,18].

Fig. 1. Deformation mechanism fields for sediments undergoing lithification. Abbreviations: I.P.F.- independent particulate flow (hydroplastic fractures plot in this field), C.P. - crystal plastic flow, D.M.T.- diffusive mass transfer, L.R.C.- low strain-rate rock creep.
If the friction mobilised within these zones is low, then they may continue to localize slip between zones of undeformed sediment. If the comminuted gouge develops greater frictional strength than the host rock, or if the normal stresses are high relative to the compressive strength of the grains, then the deformation may delocalize out of the original shear zone. In the first instance this will produce a succession of discrete, brittle cataclastic shear zones, while in the second case, which becomes more likely as confining stress increases, pervasive grain breakage may occur leading to ductile cataclastic flow [19,20].

Brittle-cataclastic behaviour is favoured in materials that have high interparticle friction, and therefore is uncommon in argillaceous sediments, or sands containing an appreciable amount of clays and ductile grains. Abrasion, wear and grain-size reduction are all observed in deformed argillaceous sediments, particularly where the clay content is less than 50%. Once fine-grained sediments are partially cemented (below), cataclasism can become an important deformation mechanism [13].

**Syn-Diagenetic and Chemically Enhanced Deformation Mechanisms**

With deeper burial, sediments tend to become more lithified though the combined action of chemical and mechanical diagenetic processes. Mechanical compaction is more important in clays than in sands, since the former have a much higher initial porosity, and are very much more compressible [5]. By contrast, the trend towards greater cementation with depth is much stronger in arenaceous sediments than in argillaceous lithologies. This is partly because of the retention of water in clays and shales, but also because diagenetic changes are impeded by reduced fluid flow in these low permeability lithologies. Typically, several tens or hundreds of pore volumes of water need to flow though a sediment to deposit one pore volume of cement. While this is possible for sands, it would typically require many of millions of years to occur in a mudrock. Also, the geochemical environments in clays may be more suitable for the mobilization of cement constituents than for their deposition (water activity is reduced and supersaturation occurs, [21]). Finally, since usually clays compact more rapidly than sands, water is expelled from them into adjacent formations (they are hydrogeochemical sources rather than sinks). The reverse may eventually occur at greater depths where the porosity of sands can be reduced to very low levels.

Deformation mechanisms occurring during diagenesis have received relatively little attention despite their importance in the evolution of many geological environments [22]. Most studies only consider the question of porosity loss in sandstones due to intergranular pressure solution [23]. Pressure solution is a type of diffusive mass transfer, whereby material is: i) removed from regions of a grain where the local free energy is high, ii) transported in solution by a fluid phase, and iii) re-precipitated in regions where the free energy is lower. The free energy gradient is commonly the result of impingement stresses of touching particles, so that material is transferred in a localized, nearly closed system. The main kinetic control on this three-step process is the activation energy of solution, so that there is an exponential increase in the effectiveness of DMT processes as the temperature increases [24]. Strictly the term “pressure solution” is a misnomer, but directed effective normal stresses control the magnitudes of the free energy gradients that drive the diffusive flow of material. Accordingly, tectonic forces can significantly enhance DMT processes.

**Fault Seal Prediction: Lessons from the Hydrocarbon Industry**

**Fault Populations, Fault Zones and Damage Zones**

Quantitative prediction of fault sealing probabilities (Fig. 2) requires an understanding of the number and spatial distribution of faults and fractures that affect the rock body under consideration. There is insufficient space to discuss this matter in any detail here. The reader is referred to reviews by Cowie et al. [25] and Knipe et al. [26]; the important points are listed below.
1. Several studies have found an approximate power-law relationship between the size of faults (in terms of length or surface area) and the number present in a given region. In other words, there are a small number of large faults and a larger number of small faults, with some characteristic sizes being favoured because of factors such as bed thickness. "Undeformed" rock units are usually characterized by a background level of faults and fractures rather than the complete absence of deformation features.

2. Faults are rarely simple planes of slip between two domains of undeformed rock. Rather slip is more usually accommodated in a fault zone consisting of anastomosing and splaying slip planes. Faults are commonly surrounded by a region of influence, where stresses associated with propagation and movement of the fault have produced a greater intensity of small faults and fractures than are found further away in the "undeformed" rock mass (Fig. 3). These damage zones may account for a considerable proportion of the overall sealing potential of a fault array. Major and minor slip surfaces may be filled with cataclastic gouge, smeared clays or precipitated minerals.

Fig. 3. (a) Fault zone as seen on seismic is a single slip plane with tip defined by resolution of the imaging. (b) Real fault zone incorporates numerous branching slip planes, a core of intense fracturing and an outer shell of moderate deformation.
3. Overall displacement is partitioned between large increments of slip on the main fault planes and smaller displacements on splays, and within minor faults in the damage zones. The details of how this slip is distributed controls the overall juxtaposition of lithologies, and so the primary sealing potential of the fault array (Fig. 4).

Fig. 4. The details of juxtaposition of permeable and less permeable lithologies (macrostructure), allied with the ability of different fault panels to seal (microstructure) controls whether horizons will retain hydraulic continuity or be sealed effectively by a fault and its damage zone.

4. It may be possible to predict the presence of small, but nevertheless important faults on the basis of a model of mechanical behaviour for a succession of rocks. This model will be conditioned by the available geological data from wells, and geophysical data from seismic lines, where only the larger faults (offset >20-30 metres) are actually resolvable. The models can also be constrained by more complete data sets from geological outcrop studies.

Microstructural Characterization of Fault Zones

The geometrical model of the fault distribution tells us only about the juxtaposition of different horizons. To date, most approaches to the prediction of fault sealing have concentrated on this problem [27]. Onto these fault surfaces, therefore, must be mapped information concerning the petrophysical properties of the fault rock produced when the two juxtaposed lithologies slip past one another. Clearly, a very large fault may be filled with a continuous zone of gouge or smeared clay, and so form an effective seal regardless of the details of the juxtaposed lithologies. However, study of numerous smaller deformation features recovered from North Sea reservoirs [26], shows that the amount of slip, beyond a few cm, is relatively unimportant in forming a sealing lithology, provided the temperature and stress are sufficient for a range of deformation mechanisms to operate.

Clay Smears

Petrophysical analyses of clay/shale smears are given by Berg [28] and Gibson [29]. Some examples of these structures are shown in figure 5, with their associated petrophysical properties. Their role in sealing high permeability units is obvious, while in low-permeability argillaceous sediments, shear zones may also have a permeability that is typically one to two orders of magnitude lower than that of the undeformed wall rocks [30,31]. Permeability reduction is caused by: i) Increased particle alignment that increases tortuosity, ii) The blocking and collapse of large sheltered pores, and most importantly, iii) An overall reduction in porosity as the result of shear-induced consolidation. This type of fault seal is probably of primary importance in argillaceous formations,
particularly at shallower levels, and since porosity is reduced and tortuosity increased across these zones, they will also act as partial barriers to the diffusive transport of dissolved species.

Fig. 5. Clay smear fault rock from northern North Sea. (a) Core specimen showing clay-rich laminae dragged across sand layers. (b) Back-scattered electron image of same rock from within smeared zone, with pore-size distribution from mercury injection.

_Cataclasites_

This is the most widely studied fault rock type, but is particularly associated with clean sandstones, silts and diatomites [17,18]. In predominantly argillaceous rock sequences, cataclasites may be important in sealing off silty layers that would otherwise be important intra-formational conduits. Some examples of these structures are shown in figure 6.
Fig. 6. Cataclastic rocks. (a) Undeformed rock with pore size distribution from mercury injection. (b) Deformed rock from within fault zone.

Cemented Fault Rocks

These fault rocks are important as indicators of past fluid activity around faults, and may be associated with embrittlement and fault breaching (see below). An example of a cemented fault is shown in figure 7. In argillaceous formations cementation may be focused along more permeable, coarser-grained horizons, and in the vicinity of faults and fractures, where thermal and geochemical gradients steepen, mineral precipitation may completely occlude fluid pathways. Thus, depending on their nature and location, cements can have a positive or negative effect on seal integrity.

Fig. 7. Back-scattered electron image of cemented fault rock from northern North Sea reservoir. The host rock is heavily cemented by carbonates but has some secondary porosity. The fault zone is completely infilled, so that porosity and permeability are reduced almost to zero. The cemented zone is fractured providing a potential conduit for fluids along the fault.
Framework Phyllosilicates

The term framework-phyllosilicate fault rock has been introduced [26] to describe an important category of fault rocks that have heterogeneous microfabrics dominated by a framework of mixed, oriented phyllosilicate plates. Although these rocks sometimes have domains of aligned phyllosilicates, these are not the well defined preferred-alignment features seen in true clay smears. Framework-phyllosilicate fault rocks can be considered transitional into clay/phyllosilicate smears as the extent and continuity of the collapsed frameworks increases with deformation.

This group of fault rocks is poorly known, and its importance in reservoir settings has only recently been appreciated [26,29]. A large proportion of most clay or shale formations actually consist of clays with less than 50% clay minerals. For these two reasons we present our findings concerning this type of fault rock in some detail (Fig. 8). Three distinct mechanisms have been identified which lead to the formation of phyllosilicate framework fault rocks in the North Sea:

(i) Deformation-induced mixing of phyllosilicate-rich lithologies. Fine grained phyllosilicates are often distributed heterogeneously throughout sandstones, along isolated laminae, within peloids or on the surface of framework grains. During deformation, the phyllosilicates become distributed more regularly, resulting in the replacement of macroporosity with a matrix of fine grained phyllosilicates. The phyllosilicates present may either be detrital or authigenic: in the latter case, the timing of deformation will be crucial to the resulting poro-perm properties of the fault rock.

(ii) Clay infiltration. Fluids passing through fault rocks during deformation have the potential to transport fine-grained phyllosilicates either by entrainment or bulk fluidization. These mechanisms require high fluid fluxes within a poorly lithified sediments, so most phyllosilicate-framework fault rocks were infiltrated by clays at an early stage, even if the deformation occurred at greater depth.

(iii) Post-deformation phyllosilicate precipitation. Many fault rocks contain higher concentrations of authigenic phyllosilicates than the surrounding host rock. For example, samples from the Brent Group in the northern North Sea contain high concentrations of disaggregated muscovite which provided ideal sites for kaolin precipitation. Therefore, kaolin preferentially precipitated within the fault rock despite the fact that the solutes for its precipitation were supplied by feldspar dissolution throughout the surrounding undeformed sediment.

Cataclasis and post-deformation pressure solution may contribute significantly towards the porosity collapse within phyllosilicate-framework fault rocks. The amount of post-deformation pressure solution increases with maximum burial depth and is an important factor in controlling the mechanical strength of phyllosilicate-framework fault rocks. Samples in which pressure solution is not pervasive may be prone to breaching as a result of fault reactivation or hydrofracturing.

Reliable permeability measurements from phyllosilicate-framework fault rocks range from $10^{-16}$ to $< 10^{-20}$ m$^2$. This represents a reduction factor of between 20 and 85000 in the background permeability of the undeformed rock. The largest permeability reduction factors appear to be associated with deformation features that have higher concentrations of phyllosilicates than the surrounding host rock. Mercury injection results yield a wide range of pore aperture sizes and threshold pressures. The higher permeability fault rocks tend to have a very broad distribution of pore aperture peaks, with moderately high average pore aperture sizes (~0.5μm). As the permeability of the fault rocks decreases, the average pore throat size decreases (down to ~0.02μm).

Fault sealing mechanisms associated with framework phyllosilicates may considerably reduce the aggregate permeability of deformed mudrocks, and so repair some of the enhancement in permeability caused by fracturing. Like clay smears, framework phyllosilicate tectonites may seal off thin high permeability horizons, but the size of the faults that act as seals in this case can be much smaller because there is now requirement for clay to be dragged across from an adjacent finer layer.
Fig. 8. Phyllosilicate framework rock from northern North Sea. (a) Undeformed rock with pore size distribution from mercury injection. (b) Deformed rock from within fault zone.

Faults as Conduits

In addition to their well understood role as seals, faults may also behave as conduits for fluid flow. Fault zones are well known as the sites of economically important mineralization, not only in “hard rock” settings [32], but also in shale units [33,34]. Hydrocarbon reservoirs are often located a considerable distance vertically and laterally from the kitchen areas in which the oil or gas was originally generated, and there are many instances where the only likely migration pathway is provided by faults. Sometimes direct evidence in the form of fluid inclusions and bitumen residues may be preserved within the fault zone [35,36], or the past presence of hydrocarbons may be betrayed by anomalous carbon isotopes in precipitated minerals. There is also evidence that currently active fault zones in muddy sediments may be the locus of focused fluid migration [31].

Mechanisms of Flow Along Mud-rich Faults and Fractures

Faults in clay and shale formations typically have a more compacted fabric than the host sediments, and so it is difficult to explain how they can behave as conduits for fluid flow unless they are dilated by some mechanism. They are also normally considered to be self-sealing, because the ductile nature of the clay enables the walls to slide past one another without generating asperities and open fractures. This is something of a misconception, because shales at depth can become brittle as they lithify, and so produce zones of fracturing that can increase overall permeability by many orders of magnitude. On a local scale, the fault itself may become embrittled by enhanced cementation or
diffusive mass transfer. More fundamentally, even un lithified clays can behave in a macroscopically brittle fashion (and dilate) if they are subjected to a shear strain under conditions of effective normal stress significantly lower than the maximum previously attained [5]. A possible example of this brittle-ductile cycling is the scaly fabric commonly found in mud diapirs and fault zones that have experienced overpressuring or reduced overburden loads during deformation [31].

Valving Behaviour

Several workers provide evidence of, and have suggested mechanisms for, episodic fluid expulsion from overpressured shale formations occurring on the basin scale [37-40]. Hunt [38] suggested that newly created fractures in shales permit flow for some time before they are sealed by cementation; Capuano [39] provided possible petrographic and geochemical evidence for the operation of this mechanism in Gulf Coast shales. Roberts and Nunn [40] reviewed a more dynamic mechanism for valving. The shales are considered to be compliant enough to keep existing fractures and faults sealed at near hydrostatic pressure. As overpressure builds (to super hydrostatic but sub-lithostatic fluid pressures) the stress differential is sufficient to initiate and hold open sub-vertical hydraulic fractures. The walls stay apart for some time (10s-100s of years) permitting rapid fluid flow, before the overpressure drops below the threshold necessary to keep the conduits open.

Valving may also occur within the fault zone itself. The fractures in a scaly clay or gouge zone are only able to act as conduits for fluid flow if they are held open by some mechanism, as normally when the surfaces are mated together, they will act to retard flow. The most likely way for this to happen in the subsurface is through an increase in the pore fluid pressure to near lithostatic values, where the normal stress and cohesion can be overcome and the shear surfaces can be exploited as hydraulic fractures [15, 30, 31].

Summary and Conclusions

In the first part of this paper we described some of the complexities that may be encountered in hydrogeological appraisal of argillaceous formations. We believe that a given formation is likely to have a highly heterogeneous distribution of porosity and permeability. Permeability may vary over several orders of magnitude at the scale of laboratory measurement, even in a rock unit that looks reasonably heterogeneous at the scale of geological mapping. It is also possible that certain lithologies have anisotropic permeability at some scales due to the presence of laminations or bedding. Structural features are likely to be heterogeneously distributed within the rock mass, with many features of hydrogeological importance being smaller that what would normally be visible at the outcrop scale.

The core of the paper describes two important aspects of sealing in fault zones. A single fault zone may consist of a number of slip planes that juxtapose different units in a complicated way. We described the various deformation mechanisms that may operate within faults and their surrounding envelope of more distributed damage. In general, deformation and associated chemical changes reduce the porosity, permeability and pore sizes of the sediments in the fault zone. Thus the fault can act as a barrier to single phase flow, diffusive flow and, particularly, to multiphase transport. In the damage zone and bend regions, fracturing and dilation may breach the fault zone and enhance fluid transport.

Finally, fault zones have been considered as possible loci for fluid flow. Experimental evidence suggests that the fabrics developed within such fault zones are not conducive to enhanced transport unless they become strongly dilated, or are embrittled and then breached by fractures. We believe that fault zones in uncemented argillaceous formations probably have sufficient ductile compliance to remain intact, with the failure planes mated together, unless they are reactivated by earth movements or hydrofractured by elevated pore fluid pressures.
Acknowledgments

This work was undertaken by the Rock Deformation Research Group of the University of Leeds. Thanks go to Greg Jones, Andy Farmer, Ned Porter, Alex Harrison, Liz White and Barbara Kidd. Support from the Natural Environment Research Council to Clennell and Knipe, and from AGIP, British Gas, BP, Conoco, Phillips, Mobil and Statoil to the RDP Group is gratefully acknowledged.

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[31] Dewhurst, et al. this volume.


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Experimental Studies on the Hydrocarbon Sealing Efficiency of Pelitic Rocks

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Abstract

Laboratory experiments and petrophysical measurements were performed to determine the hydrocarbon sealing efficiency of shales and siltstones from the Norwegian Continental Shelf and Northern Germany. The investigations covered both molecular transport (diffusion) and volume flow (Darcy flow). Diffusion coefficients for methane in water-saturated rocks measured at 150°C ranged between $10^9$ and $10^{11}$ m²/s and showed a clear correlation with TOC content. For a hypothetical scenario of a 100 m cap rock cumulative diffusive methane losses were calculated to amount to 100 up to several thousand Std. m³/m² over a period of 200 m.y. A significant reduction of diffusion coefficients was observed with increasing effective stress. Diffusion coefficients for molecular nitrogen were consistently higher than for methane.

Permeability values of the pelitic rocks varied between 0.1 and 10,000 nDarcy ($10^{-22}$ - $10^{-17}$ m²). Measurements under controlled effective stress revealed different types of stress dependence of permeability.

The effect of faults on fluid transport was investigated in a dual approach. In a microscale study permeability measurements were carried out on plugs taken in the vicinity of microfaults encountered in drill cores. In the other approach systematic measurements were carried out on pelitic rock samples from an area affected by a tectonic stress field.
Introduction

It has become an established concept in petroleum geology that hydrocarbon systems are dynamic on the geologic time scale. To improve exploration strategies the evolution of these systems needs to be understood from a qualitative and quantitative point of view. The stability of hydrocarbon accumulations and, immediately related with this issue, the sealing efficiency of barrier rocks or cap rocks is receiving increased attention. The basic concepts of hydrocarbon migration and trapping are known but only the integrated basin modelling techniques are capable of putting them to work in the complexity of natural systems.

Advanced numerical basin modelling techniques are now being used routinely by exploration geologists to analyse various hydrocarbon plays. Integration of 3D seismics, 2D and upcoming 3D models for the structural basin evolution and fluid flow provide the tools for a comprehensive modelling of geologic systems. But, these numerical tools require reliable input data in order to put realistic constraints on the modelling results.

Experimental work can, at least to some extent, attempt to provide these data and help to improve our understanding of transport processes in geological systems. The main dilemma well-known to geoscientists is the vast difference between laboratory and geologic scale both in terms of time and space. Therefore experimental work needs to be performed in close co-operation and interaction with basin modelling applications to ensure continuous feedback between the two approaches.

Concepts of hydrocarbon trapping and sealing efficiency

Separate phase fluid flow is generally viewed as the dominant transport mechanism in hydrocarbon migration starting from primary migration up to tertiary migration/dismigration. Molecular diffusion is attributed some importance mainly for natural gas dismigration and short-range redistribution of higher molecular weight hydrocarbons. Both experimental and modelling work has been conducted to achieve a realistic appreciation of the role of diffusive processes in particular in natural gas migration. Diffusive losses of gas from natural gas accumulations should not be overestimated but, as evidenced by basin modelling studies in different regions, may have a significant impact of the gas balance in hydrocarbon systems and, in some instances (i.e. stable geologic systems, long time periods, no further supply of gas) can represent a controlling factor for the depletion of reservoirs. The systematic investigation of molecular diffusion of methane in the framework of basin models has revealed the importance of dissolved methane for the gas balance of sedimentary basins.

The characteristic feature of molecular diffusion is that it occurs ubiquitously, unconditionally and permanently.

This is not the case for separate phase flow of hydrocarbons. Petroleum migration involves two- or even three-phase fluid systems and therefore capillary processes play an important role. Although conceptually well established the quantification and prediction of capillary sealing efficiency constitutes one of the major problems in the appreciation of hydrocarbon seals. For oil or gas leakage through seals to occur the capillary forces must be overcome. Only after this has happened the fluid flow will be controlled by permeability. Some of the most imminent open questions with respect to the capillary efficiency are the breakthrough conditions (breakthrough pressure, percolation threshold) for tight rocks, and the wettability of rocks and interfacial tension of the fluid phases under in-situ conditions.

Permeability is one of the key parameters in the prediction of fluid flow and pressure build-up and decline in sedimentary basins. Permeability values for tight rocks and potential hydrocarbon seals are still very limited and their measurement involves a number of experimental problems. Even if these data are available it must be kept in mind that single phase permeability is most certainly not sufficient to describe the dismigration process of hydrocarbons through seals. Future work must
address the issue of multiphase flow and relative permeabilities as a function of saturation in tight rocks.

EXPERIMENTAL EQUIPMENT AND METHODS

Based on earlier work on the diffusion of light hydrocarbons in sedimentary rocks [1], [2], [3] experimental equipment was developed for the investigation of transport processes (molecular and volume flow) in sedimentary rocks at elevated temperatures and pressures under controlled stress conditions [4]. A schematic view of the tri-axial flow cell for transport measurements in sedimentary rocks is shown in the contribution of Hanebeck et al. (this volume). This flow cell was used in various studies ranging from primary migration and petroleum expulsion to investigations of cap rock sealing efficiency. A simpler version of this type of flow cell is employed for experiments requiring no controlled axial stress.

Diffusion experiments with hydrocarbon gases have been performed at temperatures up to 180°C and fluid pressures up to 10 MPa (100 bar). Peripheral computer-controlled valve systems are used for automatic sampling of fluids from the system followed by gas analysis. As shown schematically in Figure 1 the gas diffusion experiments are performed in the absence of pressure gradients across the rock sample. Diffusion experiments have also been carried out with molecular nitrogen which represents a major component of natural gases in northern Germany.

![Diagram](image)

**Figure 1. Principle of gas diffusion measurements in water-saturated sedimentary rocks.**

Pressure-driven volume flow (Darcy flow) has a substantially higher transport efficiency than diffusive transport. Even with permeabilities at the lower end of the experimentally measurable range can significant fluid transport occur on the geologic time scale. In view of the scarcity of permeability data for tight lithologies experimental methods have been developed to measure permeabilities down to the nanodarcy range (10⁻¹⁹ m²; 10⁻¹⁴ m/s) and below. Figure 2 shows a schematic view of an arrangement used for these permeability measurements. A defined fluid pressure gradient (3-8 MPa/cm) is established across a rock slice of known thickness and the resulting flow of an incompressible fluid is measured. The flux is monitored both on the upstream and the downstream side of the sample. Experiments are carried out using a steady-state method but the flux on the upstream side is also recorded throughout the non-steady state. The confining pressure ensures a leak-tight seal around the sides of the sample plug. Experiments can be conducted under controlled stress
conditions up to a maximum axial load of 100 kN (corresponding to $= 150$ MPa for a sample diameter of 28.5 mm) and confining pressures of 50 MPa (500 bar).

![Experimental set-up for permeability measurements on tight sedimentary rocks.](image)

**Figure 2.** Experimental set-up for permeability measurements on tight sedimentary rocks.

**Application of experimental methods**

As part of a study on hydrocarbon movement and trapping pelitic rocks from the Norwegian Continental Shelf (Haltenbanken) and the North German Basin have been investigated with respect to their hydrocarbon sealing efficiency. Besides diffusion experiments with methane and permeability measurements the work comprised a geochemical and mineralogical analysis and a characterisation of the samples by standard petrophysical techniques (mercury porosimetry, specific surface area measurements by the BET method using nitrogen gas).

**Samples**

The samples used in this study were predominantly shales. In addition, two Rotliegend fanglomerate samples from a suspected lateral seal were included. Some of the Jurassic shales from the Norwegian Continental Shelf showed minor silt admixtures. TOC contents ranged up to 5% while the red Carboniferous and Permian claystones from N Germany were essentially devoid of organic matter (< 0.05% TOC). Mixed-layer clay minerals were found in traces only in the samples from Haltenbanken. Table 1 gives an overview of the ranges of mineralogical composition.
<table>
<thead>
<tr>
<th>Mineralogical composition [weight-%]</th>
<th>Haltenbanken Area</th>
<th>North German Basin</th>
</tr>
</thead>
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<tr>
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<td>16-48</td>
<td>7-30</td>
</tr>
<tr>
<td>illite</td>
<td>30-60</td>
<td>48-75</td>
</tr>
<tr>
<td>kaolinite</td>
<td>3-40</td>
<td>3-8</td>
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<td>0-10</td>
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<tr>
<td>illite/smectite-ml</td>
<td>traces</td>
<td>--</td>
</tr>
<tr>
<td>siderite</td>
<td>0-14</td>
<td>--</td>
</tr>
<tr>
<td>calcite</td>
<td>traces</td>
<td>0-12</td>
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<td>0.5-5</td>
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</tr>
<tr>
<td>TOC</td>
<td>0.3-5-5</td>
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</tr>
</tbody>
</table>

Table 1. Ranges of mineralogical composition of pelitic rocks from the Norwegian Continental Shelf and the North German Basin.

Selected results

Molecular diffusion of gases

Diffusion experiments with methane were performed at a temperature of 150°C. The methane diffusion coefficients ranged between $10^{-9}$ and $10^{-11}$ m$^2$/s (Figure 3). Conforming with earlier studies [1], [3], correlations between the diffusion coefficients and petrophysical parameters like porosity, permeability, specific surface area could not be found for the set of samples studied. Figure 3 shows, however, a clear correlation between the methane diffusion coefficients and the TOC content of the pelitic rocks. The reduction of the molecular mobility of methane in the rock samples with increasing TOC indicates an interaction (e.g. sorption or dissolution) of this compound with the organic matter. The experimental diffusion coefficients can be used to compute diffusive losses from natural gas reservoirs. To take into account the complexity of the structural and thermal evolution of sedimentary basins and the generation and migration of hydrocarbons this computation is best performed in the context of numerical basin modelling. A diffusion module was developed and incorporated into a commercial basin modelling software package (Petromod 2D of IES) to analyse the significance of diffusive transport during various stages of hydrocarbon migration. Although separate phase flow is the predominant transport mechanism during phases of intense gas generation time periods of substantial duration were found to occur where gas migration was controlled by diffusive transport [5].

To demonstrate the potential extent of diffusive losses from natural gas accumulations on the geologic time scale the experimental diffusion data for the North German shales were used to calculate diffusive gas losses for a simplified, hypothetical nonsteady-state scenario. Assuming a shale layer of 100 m thickness, initially free of gas, a reservoir depth of approximately 5000 m with a methane pressure of 50 MPa (500 bar) and a temperature of 150°C the cumulative diffusive gas losses were computed in units of Std. m$^3$ of gas per m$^2$ surface area.
Figure 3. Diffusion coefficients for methane at 150°C in pelitic rocks vs. TOC content.

Figure 4. Computed cumulative diffusive losses through different shale caprocks from N Germany.

Figure 4 shows the total diffusive gas losses through different shale lithologies over a period of 200 m.y. computed for the above scenario. The losses range between 100 and several thousand Std. m³/m².
For comparison, the average gas contents of commercial reservoirs lie between 200 and 1,500 Std. m³/m². The highest methane fluxes out of the reservoir occur during the initial nonsteady-state phase when the diffusing gas fills up the cap rock which was assumed initially free of methane. Subsequently the diffusive flux approaches a steady state value.

As stated above, experimental work on gas diffusion was also extended to molecular nitrogen. Furthermore the influence of effective stress on gas diffusion coefficients in water-saturated shales was investigated. Shown in Figure 5 are experimental cumulative diffusion curves from measurements performed with methane and nitrogen at two different values of effective stress. The increase of effective stress from 17 to 47 MPa results in a substantial reduction of the diffusive fluxes of the two gases. While the diffusion coefficients of homologous light hydrocarbons in water-saturated sedimentary rocks decrease with increasing molar mass, nitrogen (N₂), with a molar mass of 28 g/mol has higher diffusion coefficients than methane (molar mass 16 g/mol). This observation conforms qualitatively with aqueous phase methane and nitrogen diffusion coefficients reported in the literature and predicted by the Wilke-Chang relationship [6].

![Figure 5. Influence of effective stress on the diffusion of methane and nitrogen in shales.](image)

**Permeability**

Permeability measurements on the tight shale samples showed a good reproducibility (± 10% for multiple measurements on the same sample). The values determined in this study were mostly in the range from 10 to 100 nanodarcy (cf. Figure 6). Individual samples had lower permeabilities and one sample was found to be “impermeable” within the duration of the experiment so that a permeability below 0.1 nanodarcy was assumed. No correlation was found in this sample set between permeability and any other petrophysical parameters. An attempt to correlate the permeability with the measured porosity and specific surface area values according to the Kozeny-Carman relationship was equally unsuccessful.

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Figure 6. Permeability vs. porosity plot for pelitic rocks from the Norwegian Continental Shelf and the N German Basin.

Figure 7. Variation of shale permeability with effective stress.
The variation of permeability with effective stress is presently under investigation. First results obtained in this study are compared with data by Katsube et al. [7] in Figure 7. The different samples show distinct differences in stress dependence of permeability evidenced by the slopes and the shapes of the permeability - effective stress curves.

Effects of fault zones and tectonic stress fields on fluid transport

Application of the laboratory methods described above to transport in fault zones involves some serious limitations. The laboratory tests require consolidated rocks that can be machined into sample plugs of defined proportions (diameter, thickness) and orientation.

The original goal of this study was to collect core samples of shales across a fault zone and investigate the influence of faulting on the transport parameters as a function of distance from the fault plane. This approach, however, proved to be not feasible because it was impossible to get access to an appropriate fresh underground outcrop. Surface outcrops were dismissed due to weathering problems and pressure relief.

Ultimately, a dual approach was chosen to study the effect of fault zones and stress fields on fluid transport in pelitic rocks. In a “microscale” study fresh cores from faulted shale intervals were obtained from exploration wells of the German Ruhrkohle AG. Experimental work on these cores was focused on the investigation of small-scale effects (in the cm and dm range) of microfaults on the permeability of the adjacent damage zone.

Figure 8. Investigation of fluid flow in microfault systems
A schematic view of the sampling strategy is shown in Figure 8 where the locations of three sample plugs used for permeability measurements are indicated in an unrolled image of the outer surface of a drill-core section. The three plugs were taken from the same, fine-layered siltstone zone at different distances from the compressive microfault. The permeability values measured in faulted sections of

**EFFECT OF STRESS FIELDS ON FLUID TRANSPORT IN SHALES**

![Diagram showing stress fields and fluid transport in shales](image)

**Figure 9.** Investigation of the effect of stress fields on fluid transport in shales ("macroscale" approach).

this type were consistently below 3 nanodarcy (3·10^{-21} m²; hydraulic conductivity: 3·10^{-14} m/s).

The influence of tectonic stress fields on the transport properties of shales was investigated on a set of samples from a coal mine in the South Limburg mining area, The Netherlands, NW of Aachen (cf. Figure 9). Earlier studies had indicated that this area was affected by a NNW-directed stress field during the time of the Variscan orogeny. Sonic-velocity anisotropies determined on numerous orientated shale samples from coal mines in this area are attributed to this stress field. A small number of cores exhibiting distinct velocity anisotropies was selected for the laboratory studies. For the sample with the largest anisotropy (> 4500 m/s in NNW- SSE direction, < 3500 m/s in ENE-WSW direction) a strong permeability anisotropy could be established with 0.2 nDarcy in the high velocity direction and approximately 2 nDarcy in the low velocity direction.
The sonic velocity and permeability anisotropies are also reflected in the mercury porosimetry results obtained for this sample (cf. Figure 10). Porosimetry measurements were performed on 1 cm diameter plugs drilled along the directions of maximum and minimum sonic velocities.

![Graph showing capillary pressure curves from mercury porosity on two orientated plugs from a core sample of the S Limburg mining area exhibiting strong sonic velocity anisotropy.](image)

Figure 10. Capillary pressure curves from mercury porosity on two orientated plugs from a core sample of the S Limburg mining area exhibiting strong sonic velocity anisotropy.

REFERENCES.


SESSION III

Identification and Characterisation of Flow Pathways
Chairmen: J.-F. Aranyossy (ANDRA, France),
and E. Frank (HSK, Switzerland)
Timing and Pathways of Oil and Gas Leakage at Puffin Structure, Central North Sea

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Abstract

The deep Jurassic and older reservoirs of the Central Graben, North Sea, contain prolific oil, gas, and condensate accumulations. These same reservoirs have also leaked large volumes of hydrocarbons through Cretaceous seals to charge Tertiary accumulations. Symington et al. (1993) summarized studies describing the leakage paths and characterizing the products and residual hydrocarbons geochemically. This study adds new information on leakage timing to the description of the pathway from the Jurassic-Triassic reservoirs in the Puffin structure. Timing the leakage permits a more complete evaluation of the pressure, temperature, and effective stress conditions in the seal during failure.

In this study the burial evolution of the on- and off-structure portions of the Puffin sub-basin is tied to the diagenetic history inferred from the rock microstructures. Analyses of water and hydrocarbon filled fluid inclusions provide the link between diagenesis and the initial migration of hydrocarbons into the structure. Lastly, dating of the late stage clay formation, within the reservoir and above the paleo-oil water contact, constrains the timing of the leakage from the trap. Our interpretation is that clay mineralization occurs through re-saturation by aquifer waters in one fault-sealed portion of the trap. We interpret present-day profiles of pressure in the hydrocarbon and water legs of the segmented trap to imply continuity within the hydrocarbon leg above discontinuous aquifers. When we tie the present-day profiles of temperature and rock maturity to maturation models, we can infer the timing of leakage. The latest burial episode is the dominant heating event, both on structure and off. Reservoir rock and fluid observations fit an impulsive leakage event between 10 and 4 ma (although minimum duration of the episode may be less than 6 ma).

We mapped the portion of the seal interval along the leakage pathway within the 3D seismic survey. Our interpretation outlines a zone of disrupted seismic reflection amplitudes, within the sealing Cretaceous clays and marls. This zone widens upward from a limited entry area at the base of the Cretaceous seal located over a complex set of underlying faults in the Jurassic and basement rocks. Possible parameters of hydraulic conductivity for the pathway will be discussed with the limited rock sample observations. Puffin represents a specific instance of seal failure allowing rates of fluid movement to be computed. Thus, at least in this area, leakage of oil and gas is shown to be an effective means of charging the overlying Tertiary reservoirs.
Fracture Control of the Hydrology of the North American Midcontinental Cretaceous Shales

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The Cretaceous shale section of mid North America, which reaches more than 2 km in thickness and two million km$^2$ in areal extent, is among the largest accumulations of low-permeability argillaceous sediment in the world (Figure 1). As such, it is of particular interest to understand the architecture of the shale's permeability and the role of fractures in controlling its hydrology. Fortuitously, several independent studies conducted within the last 15 years have supplied pieces of the puzzle. Bredehoeft et al. [1], Case [2], and Butler [3] analyzed the regional groundwater flow systems in South Dakota and North Dakota, Belitz and Bredehoeft [4] analyzed the regional groundwater flow system in the Denver Basin region, and Neuzil [5] delineated the groundwater flow regime within the shale itself at a site (Hayes) in South Dakota approximately one km$^2$ in area. Synthesis of the results of these studies suggests that fractures play an important role in the hydraulic behavior of the confining layer and control how it interacts with aquifers.

**Permeability of intact shale.** The intrinsic permeability of the Cretaceous shale (that of the intact shale material) has been reasonably well established from cores and borehole tests in South Dakota and Nebraska. The core and borehole data were obtained using several different techniques, but are quite consistent. They point to permeabilities in the range $10^{-21}$ to $10^{-20}$ m$^2$ with some evidence for both higher and lower values [5] [6]. The range in values is probably due to slight lithologic variations, differences in compressive load with depth, and the difficulty of measuring such small permeabilities.

Although the core and borehole permeability data are not well distributed over the extent of the confining layer, other evidence indicates that the intact shale permeability is quite consistent everywhere. The additional evidence comes from larger scales of investigation in areas that do not show evidence of secondary permeability.

The Hayes site study [5] showed no indication of secondary permeability. This study examined conditions in the shale directly, rather than through conditions in the aquifers as in other studies considered here and was unusually well constrained. It revealed a flow regime marked by a significant hydraulic head minimum within the shale and flow into the shale from both upper and lower boundaries. The vertical hydraulic gradients driving flow toward the shale interior are large (on the order of unity), but no horizontal hydraulic gradients were observed. It seems clear that this head pattern represents a hydrodynamic disequilibrium related to elastic rebound of the shale after erosion of younger strata [5]. One can think of this system as analogous to a large-scale permeability experiment, in this case a transient consolidation/rebound (oedometer) type of procedure. An analysis that is similar to that for oedometer testing yielded an estimate for the shale's permeability of $10^{-21}$ to $10^{-20}$ m$^2$. This is comparable to laboratory and in situ values of shale permeability and indicates an absence of
secondary permeability at the study site. In addition, the absence of horizontal hydraulic gradients implies that transmissive fractures cannot be nearby, as they would disrupt, or "short-circuit" the disequilibrium.

![Map of North America with regions marked]

**Figure 1.** The Cretaceous shale confining layer of the North American midcontinent. Regions labelled 1 and 2 and the intervening area marked by dashed lines show the approximate extent of the shale. The shale's regional permeability is enhanced by fractures in the northern portion of region 1 (South Dakota) as well as in North Dakota (state enclosing D'). Elsewhere, fracture permeability is not evident. Regions labelled 3 show pre-Cretaceous shales that are not discussed here, and D-D' denotes the location of the section shown in Figure 3. Reproduced from [11].

An analysis of the confining layer in the Denver Basin also showed no evidence of secondary permeability. The Denver Basin is marked by strongly subhydrostatic fluid pressures in the Dakota Aquifer, a regional sandstone aquifer below the Cretaceous shales (Figure 2). Different mechanisms have been proposed to explain the low pressures. Ottman [7], for example, argued that they represented a hydrodynamic disequilibrium caused by erosional
unloading. In contrast, Belitz and Bredehoeft [4] analyzed regional groundwater flow in the Denver Basin region as steady-state and topographically-driven. One can think of the system envisioned by Belitz and Bredehoeft as analogous to a large-scale steady-state permeability experiment. By adjusting the regional vertical permeability of the Cretaceous shales, they were able to mimic the hydraulic heads in the underlying Dakota aquifer (Figure 2). They found that their simulations required a single shale permeability of approximately $3 \times 10^{-20}$ m$^2$ or less or shale permeabilities that varied between $10^{-19}$ and $10^{-21}$ m$^2$ depending upon depth. Like the permeabilities obtained in the Hayes study this range agrees remarkably well with laboratory and in situ permeability values for the intact shale, and thereby indicates an absence of secondary permeability. This is a provocative result because it shows no permeability increase with scale in the shales over a surprisingly large region.

![Figure 2. West to east cross-section through the Denver Basin. The stipple and block pattern indicate arenaceous and carbonate aquifers below and within the Cretaceous shales. The dashed line is the hydraulic head in the Dakota Sandstone; the water table is too close to the surface to be shown. Reproduced from [11].](image)

**Evidence for secondary permeability in the confining layer.** Analyses of regional flow in the Dakota Aquifer in South Dakota [1,2] and North Dakota [3] provide contrasting results; these studies indicate much higher regional permeabilities for the Cretaceous shales. Like the Dakota Basin study [4], they treated the flow system as an equilibrated, topographically-driven one, and like the Denver Basin study, they were analogous to steady-state permeability experiments in the laboratory. In each, it was possible to mimic the hydraulic heads in the Dakota aquifer only if the shale permeability is between $10^{-18}$ and $10^{-16}$ m$^2$. These results clearly indicate the presence of secondary permeability, because the regional permeabilities are two to five orders of magnitude larger than the intact shale permeability. It is also worth noting that the study by Bredehoeft et al. [1] estimated regional permeabilities for each three of the shales in the confining layer. They found a trend of decreasing regional permeability
with increasing shale depth. This is entirely consistent with secondary fracture permeability that is reduced as the fractures squeeze closed with increasing compressive stress.

Figure 3. North to south cross-section of the Cretaceous shales and underlying Dakota Sandstone showing regional vertical permeabilities of the shales. (a) The permeabilities obtained from regional flow analyses and interpreted as pertaining to geographic areas or facies of the shale. The shaded and blank portions of the shale simply correspond to the analyses shown at the top of the illustration. (b) The conceptual model presented in this paper. Here, the shaded and blank portions of the shale indicate depths of less than and more than one kilometer, respectively. Reproduced from [11].

Numerous minor, off-vertical fractures in the shales have been observed in boreholes and cores, but analyses of numerous borehole responses indicate that they are not transmissive.[5]. These minor closed fractures may form a dense network between larger, transmissive fractures. Such a pattern is reminiscent of fractures in crystalline rocks, where sparse, highly transmissive fracture zones are often connected by much smaller fractures. In the shale, however, the minor fractures leave the shale hydraulically intact.

The hypothesized transmissive fractures have never been observed, although additional geochemical, geothermal, and geomorphological evidence points to their existence [8] [9] [10]. They may, for example, be related to northwest-southeast linear features in the topography. Such features can be observed in digital relief maps (see, for example, Thelin and Pike, [10]) to extend from the Mexican border northward across the U.S. Great Plains and into Canada. Thus they seem to be structural features related to regional processes, such as basement tectonics.
Unifying conceptual model. The studies we have described suggest widely different shale permeabilities and flow regimes and, at first glance, appear to be mutually inconsistent. While it is possible that one or more of the analyses is flawed, it is also possible to reconcile the results in a single conceptual model of the shale hydrology. Transmissive vertical to subvertical fractures or similar features are a central feature of such a model. We hypothesize that such transmissive fractures control the regional vertical permeability of (and leakage through) the shales, but only at depths of less than approximately a kilometer. This aspect of the model is shown in Figure 3. This explains the significant leakage fluxes through the shales reported in the Dakotas, while allowing the smaller intrinsic shale permeability to control regional behavior of the shales in the Denver Basin. This results from the fact that the base of the shales is deeper than one kilometer over much of the Denver basin, while it is shallower than one kilometer over much of North Dakota and South Dakota.

To complete the conceptual model, we must place a further constraint on the fractures. The site study in South Dakota found no evidence of the vertical transmissive fractures. The nature of the study suggests the existence of a block of “tight” shale that is at least a kilometer across. We hypothesize that such blocks are characteristic. In other words, the fractures are very sparse and separated horizontally by a kilometer or more.

The different regional leakage rates in different shale units reported by Bredehoeft et al. [1] imply that the vertical permeability decreases by three orders of magnitude between the near surface and one kilometer depth. If one assumes single vertical fractures (rather than fracture zones) are present and if a simple parallel plate model is used to compute flow, this corresponds to an order of magnitude decrease in fracture aperture. If one accepts a one to two kilometer spacing for the fractures, the regional permeabilities in South and North Dakota imply fracture apertures that are about 100 microns near the surface and that are squeezed to about 10 microns as a depth of one kilometer is approached. This may be compared with a representative pore size in the intact shales of about one micron. The absence of secondary permeability below approximately one kilometer depth implies that the 10 micron fracture openings are lost with further increases in effective stress.

One is left with the apparent inconsistency of regional flow analyses that treat the flow system as an equilibrated, topographically-driven one, while we hypothesize significant disequilibrium conditions in the bulk of the shale. Simulations show that the disequilibrium fluxes into the “tight” shale blocks are much smaller than the fluxes through either the aquifers or the fractures. We suggest that this has permitted development of a “hybrid” flow regime, with disequilibrium conditions (caused by erosional unloading) within the large blocks of “tight” or hydraulically intact shale, and largely equilibrated flow through the transmissive fractures and aquifers. The term “hybrid” refers to the fact that characteristics of both equilibrium and disequilibrium can be observed in the flow system, depending upon the scale and type of observations one makes.

It is also possible that there are no transmissive fractures in the shales at all, and that the earlier studies must be reinterpreted. In either case, the shale section is an island of relatively low permeability in mid North America and demonstrates that heterogeneity of permeability in the shallow crust occurs at scales up to thousands of kilometers. However, within this low-permeability island the permeability, as well as the apparent type of flow regime, depend upon the scale and nature of one’s observations. The vast bulk of the shale appears to be controlled by the intact shale permeability, which is quite small. As a result chemical mass within the shale is probably sequestered for geologically significant periods of
time. This has important implications for the isolation of toxic materials.

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The Secondary Permeability of Italian Clays: A Review

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Abstract

Over the years several studies have been performed in Italy on the permeability of various argillaceous formations for the purpose of assessing their potential utilization for the isolation of long-lived radioactive waste. The main organizations involved in the studies are Ismes, ENEA and a variety of university groups, among which it is worth mentioning the soil gas team at the University of Rome, that has acquired a good knowledge of the transport of gas through a variety of geological materials, including clays.

During the 80s, Ismes, under contract with ENEA, has performed an extensive survey of tunnels intersecting clay formations for the purpose of identifying water inflows and of interpreting them in relation to the nature of the water-bearing features present outside the lining.

In the same years additional investigations have been carried out on the relevance of oxidation haloes observed around fractures in many Italian clay quarries.

In the late 80s, Ismes has participated to the "Faults in Clays" project, partially financed by EC. The main objective of this project was to improve the sensitivity and resolution of geophysical techniques for identifying and characterizing faults intersecting clay strata. In the course of this work some additional observations on the in situ permeability of the investigated clay formations were effected.

All these observations lead to a somewhat complex picture of fluid flow through argillaceous materials. The first obvious conclusion is that generalizations are not possible: argillaceous formations are characterized by extreme variability in respect to intrinsic properties, sedimentological and structural set-up, consolidation history and regional stress conditions. As a result of this complexity widely different permeability, for both gas and water, has been observed even in apparently similar materials. In addition gas data indicate that flow, at a particular location, can vary also as a function of time.

At this point the only prudent approach would seem to require that every clay formation or potential site is assessed on its own merits. For the characterization of specific locations, in respect to potential fluid flow, a variety of techniques and procedures are now available; these include: high resolution geophysical techniques, in situ testing, field surveys and soil gas analyses.

In the meantime, it is important that field observations of fluid flow through clay-rich materials continue to be performed. An essential task would be to correlate flow data with boundary conditions; primarily in order to clarify the role of the stress field as a controlling factor of fluid flow in specific structural features.

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1. INTRODUCTION

The Italian programme on disposal of long-lived radioactive waste has always been involved with the assessment of argillaceous sediments as potential host rocks of waste repositories. Argillaceous sediments are very abundant in Italy. Large accumulations of pelitic materials have taken place in connection with the geological events due to the Alpine orogenesis. Following the uplift of the Apennines, that occurred mostly in Miocene time, significant thicknesses of clays were accumulated in the valleys and in the foreland. These clays, accumulated in Pliocene-Pleistocene time, are usually characterized by a number of common features, regardless of their specific location in the Italian peninsula. In general the Plio-Pleistocene clays of Italy are bluish grey in colour, relatively rich in calcium carbonate and moderately stiff. While these clays, called in this paper “blue clays”, are not the only argillaceous formation that could be considered for disposal of radioactive waste in Italy, they have been the object of most investigations and, therefore, are the best known material in respect to isolation capacity.

Older clays are also well represented; they can be grouped in allochthonous clays, that have been emplaced by gravitational sliding associated with the uplift of the mountain chain, and in other pre-pliocenic clays. The horizontal displacement of the allochthonous formations is rather variable, but in some cases it can be as much as hundreds of kilometers. The typical aspect of the allochthonous clays is multicoloured masses of scaly, indurated clays. As a result of the orogenic transport the clays are highly tectonised and occasionally include large blocks of other rocks that were thorn from the substratum. These clays are known by a variety of local names but the most usual and the one used here is “Argille Scagliose” (scaly clays). The age of the Argille Scagliose vary from Cretaceous to Oligocene.

The other pre-pliocenic clays include all remaining argillaceous formations in Italy. Most of these clays are miocene in age and the parautochthonous units are strongly tectonised, although from the granulometric, mineralogical and hydraulic viewpoints they can be quite homogeneous. The flysch deposits, on the other hand, are rather heterogeneous and can contain complex water channels. Clays outcrop over large areas of Italy, but their direct observation is usually prevented by the presence of soil and vegetation; consequently available information about a variety of features that can be important in respect to their capacity to perform as a barrier is derived from exposures in quarries and road cuts and from tunnelling work. In addition there are additional useful indications that can be obtained from hydrogeological interpretations and, in recent years, from waste-related work and soil gas studies.

Over the past twenty years clays have been investigated, in the framework of radioactive waste disposal programmes, for various purposes and at different locations. Unless specified otherwise the following activities were performed by Ismes on behalf of Enea in the framework of contracts between EC and Enea. A non-exhaustive list of investigations addressing some aspect of the secondary permeability of clay-rich formations is as follows:

- assessment of quarries and tunnels for the purpose of identifying a suitable location for an underground laboratory; this work was performed in the early 80s;
- overview of literature about geotechnical properties, water occurrences and permeability of clays;
- observations in quarries and road cuts;
- observations in tunnels, either directly or collecting information from a variety of subjects through interviews and questionnaires;

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• measurements in quarries to reveal any alterations undergone by clays in proximity to faults and joints (performed by Ismes on behalf of JRC, Ispra);
• research on the detection of faults in clays by means of geophysical techniques - Faults in Clays Project (performed by Ismes, BGS and Exeter University with the financial support of EC and national organizations);
• soil gas studies to reveal the gas permeability of several clay formations in Italy and UK (performed by Exeter and Rome Universities with the assistance of Ismes and the financial support of EC).

2. OBSERVATIONS

The observations discussed below are divided on the basis of geological formation and of investigation procedure.

2.1 Flysch Formations

A literature survey and interviews with tunneling experts have provided some information about water circulation in two flysch formations: the Laga formation and the Numidian flysch [Chiantore and Gera, 1986].

The *Laga formation*, probably of Miocene age, outcrops extensively in central Italy, on the Adriatic slope of the Apennines. It consists of large amounts of argillaceous-silty sediments interbedded with sandy layers of variable thickness. Several tunnels of the motorways Roma-L’Aquila and Roma-Pescara have been excavated in this formation.

• **Carrito gallery** - This gallery, excavated in the stretch Avezzano-Pescara, crosses the Laga formation in its more argillaceous facies. At this site the flysch is overthrust by a paleogenetic, water-bearing formation of marly limestone. The area is interested by tectonic dislocations (fractures and faults), many of which have been intersected by the gallery. Apparently the discontinuities do not cause any visible alteration of the flysch neither in respect to geotechnical properties nor to water percolation. As a matter of fact the Carrito gallery, up to the contact clay-limestone, where the overburden is approximately 300 m, has been recorded as a completely dry excavation.

• **Gran Sasso tunnel** - This double tunnel is over 10 km long and more than 3 km are excavated in the Laga formation. The flysch, overlying a limestone of upper Miocene age, is overthrust by Mesozoic limestones. Marly clays in layers of thickness ranging between a few decimeters and a couple of meters make up most of the flysch. Occasionally some levels are silty or sandy. The marly clays are well consolidated and irregularly fractured. Many fractures are filled by secondary calcite. The content of calcium carbonate is variable but always high (35 to 65%). The intensity of fracturing is variable and related to the proximity to tectonic dislocations. Thus near the Mulattieri fault, tectonic contact with the Miocene limestone, the marly clays appear extremely fractured, divided in shiny scales, with the layers no more recognizable. Where the tunnel is overlain by the thickest overburden, the overlying water column is estimated to reach about 600 m with a hydrostatic pressure over 60 atm; nevertheless tunneling proceeded without difficulties and water inflow was restricted to localized seepages.

The *Numidian flysch* consists of alternating pelites and arenites, with a maximum thickness around 2000 m. The main outcrops are in northern Sicily. The formation was accumulated by turbidity flows in a period going from Oligocene to lower Miocene. The Numidian flysch is extremely
heterogeneous; the upper part of the formation is often cahotic, which is interpreted as an indication of the partially allochthonous nature of the material. The Numidian flysch has witnessed several tectonic events related to the tecto-genesis of the Apennine-Maghreb system. Several engineering works located in Numidian flysch have caused geotechnical investigations and detailed geological surveys. In general some zones can be recognized in Numidian flysch on the basis of the weathering level and of the intensity of fracturing. The colour varies from yellowish gray near the surface to dark gray at depths greater than ten meters. Fractures are widespread, delimiting blocks of some dm$^3$ in volume at depth and increasing markedly in frequency near the surface. On the other hand the intense fracturing of the formation does not result consistently in the enhancement of the permeability, which appears to suggest that, at times, the overburden pressure is able to keep the fissures tightly closed. Field observations support this interpretation.

- **Scanzano and Rossella dams** - These two dams, located on Numidian flysch near Palermo, have been thoroughly monitored for a number of years showing no loss of water due to underground seepage.
- **Bifarera-Scanzano tunnel** - During the excavation of this tunnel no water percolation was observed, with the exception of a water seepage due to a sand layer.
- **Crisa gallery** - This is an hydraulic gallery near Enna. The Numidian flysch at the site is argillaceous and scaly, with some thin intercalations of sands. The formation includes also some quartz-arenitic olistoliths with dimensions of a few tens of meters. The whole area shows signs of intense tectonic activity and folds, faults and fractures were encountered during excavation of the gallery. Tunnelling work was occasionally complicated by rock instability due to swelling in proximity to discontinuities made evident by stress release. In some cases, particularly near arenitic levels, water and methane entered the gallery. It is worth mentioning that in most cases the water inflow was not immediate, but seepage started with a delay and dampness of the shotcrete could be observed only some days later. A possible interpretation of this delay is that the discontinuities are normally tight and only after relaxation they are able to transmit fluids. The fluid reservoirs are probably arenitic lenses intersected by the fractures at some distance from the gallery.
- **Risalaimi landslide** - This landslide near Palermo interests Numidian flysch. Pumping tests at the site have shown the existence of water percolation at the contact between the unaltered deep clay and the weathered surface layer. In addition water seepage could be observed within relaxed fractures in the clay outside the slide area.

### 2.2 Miocene Clays

Miocene clays exist in many parts of Italy. An observation performed in a Sicilian occurrence is described below.

- **Disueri gallery** - This gallery, located near Gela, is the outflow of the lake of Disueri. It is excavated in argillaceous sediments of Tortonian age (upper Miocene), that are not part of Numidian flysch. The formation is considered to be autochthonous and is relatively free of tectonic disturbances. The gallery is 400 m long and the maximum overburden is about 60 m. It was excavated with the lake already full of water (maximum head between 10 and 15 m). As the excavation approached the lake the sediments became progressively more marly to change eventually into a diatomitic marl, called Tripoli, which was overlain by the limestone forming the bottom of the lake. The excavation was completely dry until the limestone was reached. It is worth
mentioning that the argillaceous formation is layered, overconsolidated, fissured and under a limited lithostatic load; nevertheless it appears to be completely impermeable. From a geotechnical point of view the formation was very stable; as a matter of fact no convergence of the gallery walls was observed and emplacement of the lining could be delayed until several months after excavation. No water percolation was noticed during the period when the gallery remained unlined.

2.3 Allochthonous Formations (Argille Scaglione)

Argille Scaglione are widespread in the Italian peninsula (particularly in the south) and in Sicily. Their age of deposition varies between Mesozoic (usually Cretaceous, occasionally older) and Oligocene, but their gravitational sliding, away from the uplifted sedimentation basins, has taken place much later, in the framework of the uplift of the Apennines.

Different units of Argille Scaglione can differ significantly in respect to mineralogical composition, extent of scaliness, amount and dimensions of inclusions. Usually, outcrops of Argille Scaglione are characterized by marked slope instability and widespread gravitational sliding. In addition to small scale, shallow movements, large masses of Argille Scaglione can be affected by deep seated, long-term movements that seem to indicate the continuing migration of these masses. Despite their scalyc structure and the extremely high level of tectonic disturbance, Argille Scaglione are characterized by very low permeability; both in situ and laboratory determinations have given values of hydraulic conductivity below $10^{-11}$ m s$^{-1}$ [D'Elia, 1977].

Some field observations are described below.

- **San Donato tunnel** - This is a railroad tunnel on the Rome-Florence line, not far from Florence. The tunnel is long about 10 km, and around 4 km are in Argille Scaglione. Despite the presence of a number of faults no water seepage was observed in the section in clays. The only problems with water were due to drainage from an adjacent aquifer when the tunnel slope carried the water within the clays.

- **Ogliastro gallery** - This is a hydraulic tunnel excavated near Raddusa in Sicily. The clay formation is a local variety of allochthonous argillaceous complex called in the area “Argille Brecciate”. Their age is somewhat younger than typical Argille Scaglione (middle-upper Miocene). The formation consists of marly and well consolidated clay fragments dispersed in an argillaceous matrix. The formation is intersected by fractures and the gallery crossed a fault at a depth of about 150 m. The fault is accompanied by a sizable zone of tectonic breccia that comprises blocks of different rocks. When the gallery entered the fault zone blocks of limestone, sandstone and gypsum collapsed in the opening. An inflow of water containing sulphur compounds, probably derived from the overlying “gessoso-solfifera” formation (an evaporitic complex containing diatomitic marls, limestone and gypsum), occurred as well. The water inflow was initially about 15 Ls$^{-1}$ and then decreased progressively to about 3 Ls$^{-1}$; there is little doubt that the fault zone intersected by the Ogliastro gallery is rather permeable.
2.3 Blue Clays

2.3.1 Tunnels

The blue clays of Plio-Pleistocene age are widespread on both sides of the Apennine chain and in Sicily. They are characterized by significant variability in respect to mineralogical composition, grain size distribution and carbonate content. The average content of clay minerals is around 50%, the rest consisting of silt and carbonates. Generally they are characterized by a certain level of overconsolidation, which, at least partially, could be apparent and due to cementation by the carbonates.

The blue clays were accumulated in internal basins, formed during the pliocenic marine transgression, and in the coastal and pelagic environments surrounding the Italian peninsula. Many of the internal basins evolved into brackish and fresh water lakes. Thus some blue clay deposits have been accumulated in fresh water environments and usually contain large amounts of organic matter and, occasionally, beds of peat or lignite. The blue clays of marine origin are often interbedded with thin sand levels, particularly in proximity to the pliocenic coasts. Their mineralogical composition reflects the composition of the parent rocks.

Some blue clay formations are parautochthonous, that is they were accumulated on a moving allochthonous unit. It is likely that the movement of the substratum has exposed the accumulating sediments to significant stress and possible fracturing. Regardless of the mechanism, most blue clays are fissured, the block jointing affecting the entire body of the unit and not only the near-surface strata. As a matter of fact tunnelling work in blue clays confirms the widespread occurrence of more or less latent fissures.

During construction of the new railroad line Rome-Florence several galleries were excavated in blue clays. Some observations are described below.

- **Castiglione in Teverina gallery** - This gallery, almost entirely excavated in clays, under a maximum overburden of about 160 m, is 7.5 km long. During the tunnelling work, the rock manifested two different behaviours: at times the clay had properties similar to a marl, with small scale conchoidal fractures, elsewhere the clay behaved more plastically and broke in regular blocks. No visible differences existed between the two materials, thus it was hypothesized that the difference in behaviour was due to variations in water content. The gallery encountered major discontinuities every 30 to 50 meters and sometimes rock falls occurred. Occasionally the rock falls exposed smooth and shiny surfaces, indicating that the discontinuities were real faults. However no water inflow was observed, neither in the intact rock nor along the faults.

- **Orte gallery** - On the contrary excavation of this gallery, located not far from the previous one, was complicated by water inflows and a variety of stability problems. The stratigraphic situation of the two galleries is the same, but the morphological features are different. At Castiglione, contrary to Orte, relief is significant, so that drainage of meteoric water should be much faster with a reduced infiltration.

- **Aurelia gallery** - This is one of several galleries excavated through pliocenic clays within the Rome city limits. This specific clay formation is known locally as “Argille Vaticane” and can reach significant thicknesses; 700 m were crossed by a borehole drilled at Circo Massimo in 1934. The Aurelia gallery intersects the near-surface section of the formation where thin sand layers interbedded with the clay are rather abundant. Many joints and fissures are met by the gallery, some of them are water bearing. But
there is a suspicion that the boreholes drilled during the site investigation may have created connections between the overlying aquifer and the sand levels, increasing the water seepage.

- **Sarmento tunnel** - This is a hydraulic tunnel located in Lucania. Southern Italy has been affected by intense tectonic activity that in many cases is still going on; therefore the blue clay formations must be intersected by many dislocations. The Sarmento tunnel passes under a surface cut that is generally interpreted as of tectonic origin. When the tunnel intersected the underground projection of the incision, no water inflow nor stability problems were observed. However four to five months after the excavation, swelling of the clay, water seepage and lining deformation took place. Water inflow occurred both along sandy layers and through fissures. The sequence of events was interpreted to mean that the stress release due to the excavation caused the fissures to relax and caused a significant secondary permeability in a previously tight formation.

It would be possible to continue with the description of clay behaviour in tunnels, but the examples described above may be enough to confirm the great variability of hydraulic properties of blue clays. Table 1 summarizes the observations on water percolation in Italian clays up to 1988 [Gera and Chapman, 1988].

### 2.3.2 Clay Alterations near Faults - Oxidation Haloes in Quarries

In the framework of a study that Ismes performed on behalf of the JRC, Ispra, in the early 80s, detailed observations were carried out in some blue clay quarries in central Italy [D’Alessandro and Gera, 1986].

The main thrust of the study was two fold: sampling and testing clay samples around faults to identify any fault-related alteration and observation of oxidation zones that occasionally accompany fissures and joints in clay quarries.

- **The Orte quarry** - The clay at Orte is a typical blue clay. The silt and carbonate content is relatively high; clay minerals make up 50 to 60% of the rock. Porosity, and thus water content, is about 20%. At the time of the study the quarry front was about 300 m long; the maximum eight, about 90 m, was broken in some steps. Several systems of fractures and fissures could be observed including a very obvious (at the time) normal fault with about one meter of throw. The fault plane was shiny and did not show any sign of water percolation. On the contrary, oxidation zones were well developed around many fissures exposed in the quarry. Clay samples were taken in proximity to the fault plane on a step about 60 m below the original ground surface. Sampling covered an area going from the fault to about 10 m away. No variation of clay properties, that could be interpreted as resulting from the presence of the fault, was identified. Also analyses performed on coloured material obtained from the oxidation zones did not show any significant alteration, with the exception of a slight enrichment of emathite and lepidocrocite which explains the colour difference. Permeability measurements of fissured and un fissured samples (under confining stress) have given hydraulic conductivity values ranging between $2.6 \times 10^{-11}$ and $2.2 \times 10^{-11}$ m·s$^{-1}$, indicating that “closed” fissures of this type have a negligible effect on permeability. On the other hand the presence of the oxidation zones proves that, at some time, water has circulated through the fissures. Obvious questions can be asked about the time of water circulation and the rate of formation of the oxidation zones. Two explanations are possible:
1) the oxidation zones are formed over geologic time within the clay mass; in this case they would provide some evidence about water circulation in clay;
2) the oxidation zones are formed rapidly and very close to the surface, probably as a result of fissure formation by drying and shrinking; in this case the oxidation zones visible in the lower levels of the Orte quarry would be subsequent to quarrying; activity that started a little over hundred years ago.

In the second case the oxidation zones would not represent an evidence of long-term water percolation.

- **The Santa Barbara mine.** A lignite deposit has been strip mined at this site. The lignite was located at the base of a fresh water clay-rich sequence of pliocenic age. The upper section of the lacustrine complex is formed prevalently by clays with small interbeds of sand and lignite. The Santa Barbara clays are overconsolidated, stiff and intersected by an unusual number of fissures and joints. Some true faults are also present; in some cases the faults are surrounded by a thickness (up to a few meters) of intensely tectonised rock. Sampling in proximity to a fault plane was performed at Santa Barbara as well; even in this case no fault-related alteration of the intrinsic clay properties was identified. On the other hand a quick survey of the mine provided some useful information. In the first place the Santa Barbara clays are characterized by a non-negligible secondary permeability, as proven by the existence of a water table in the formation. A number of wells were permanently pumped at the time to lower the water table in the mine area. Secondly, in many locations in the mine, clay blocks were coated with a thin layer of reddish-brown material. The phenomenon at Santa Barbara is obviously different from the oxidation zones observed at Orte. As a matter of fact at Santa Barbara the coloured coating is attached to the clay blocks without diffusion in the clay matrix. Clay blocks with the reddish-brown coating are particularly common in the higher levels of the mine, but, in one particular case, coated blocks have been found in the lowest section of the pit, about 120 meters below the original ground surface. Unfortunately no information exists about the existence of the coloured coatings before excavation of the mine.

Unfortunately, the geotechnical, mineralogical and chemical determinations failed to reveal any fault-related alteration of clay properties.

Some useful indication can be deduced from the observations carried out with the Scanning Electron Microscope (SEM). The bulk clay at depth presents an anisotropic microtexture; it also appears wet and overconsolidated. When the clay is brought to the surface, drying and decompression take place, resulting in microtextural changes. The most important modification consists in the formation of fracturing surfaces, where the mineral particles rearrange themselves in a more isotropic way.

Assuming that similar phenomena take place in situ, the microfractures generated by decompression and shrinking at shallow depth would allow iron-rich, oxidizing solutions to percolate, leading to the formation of coloured coatings or oxidation zones, depending on the specific conditions of the site.
Table 1. Observations of water percolation in Italian clays (Gera and Chapman, 1988).

<table>
<thead>
<tr>
<th>Observations No.</th>
<th>Type</th>
<th>Discontinuities recognized</th>
<th>Water inflows</th>
<th>Depth (m)</th>
<th>Notes (suspected causes of fracturing and water seepage, etc.)</th>
</tr>
</thead>
</table>
| PLIO-PLEISTOCENE BLUE CLAYS
| 13               | Literature*       | 10                         | 2 (?)         | up to 100  | Stress release near surface; permeability anisotropy due to sedimentary features; local variations in subsidence rate. |
| 11               | Quarries          | 10                         | 1 (?)         | 10 to 60   | Difficult to distinguish true faults from joints and fractures; almost impossible to see water percolation. |
| 16               | Tunnels (visited or reported) | 13 | 9 | 100 to 300 | Stress release at low overburden; silt and sand lenses connected to aquifers; discontinuities as surfaces of deep slope movements. |
| 40               |                    | 33                         | 12 (?)        |           | (Totals)                                                     |
| PRE-PLIOCENE CLAYS AND PELITIC FLYSCH
| 6                | Literature        | 4                          | 1 (?)         | ?         | Stress release near surface; water percolation in fissure network of weathered superficial strata. |
| 9                | Tunnels           | 8                          | 5             | 15 to 100 | Coarse-grained layers; very high tectonization up to pseudo scaly type. |
| 15               |                    | 12                         | 6 (?)         |           | (Totals)                                                     |
| ARGILLES SCAGLIOSE (SCALY CLAYS)
| 6                | Literature        | 3                          | -             | ?         | It is very difficult to recognize faults in this material. |
| 9                | Tunnels           | 9                          | 4             | 80 to 350 | Discontinuities as slip surfaces of gravitational movements; breaks after earthquakes. |
| 15               |                    | 12                         | 4             |           | (Totals)                                                     |

* Mainly surface observations and geotechnical determinations.

2.3.3  Faults in Clays Project

The Faults in Clays project, partially financed by the European Commission, was aimed at assessing the possibility of employing non-destructive techniques for the detection of faults in argillaceous formations (Gera et al., 1991). The project was a joint undertaking of BGS and Exeter University in UK and of Ismes and ENEA in Italy. Specific objectives of the project were:

- to assess the capability of geophysics to detect faults in clays;
- to measure directly the hydraulic properties of fault zones;
- to define the site investigation approaches for locations where faults are to be expected.

Relatively different approaches to the project were taken in UK and Italy, mainly owing to the differences between the investigated clays in the two countries.

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**Down Ampney Investigations**

The British site was located at Down Ampney where Oxford clay is intersected by a normal fault with a throw of almost fifty meters (Gera et al., 1991). Geoelectrical techniques were successful in revealing the presence of the fault, even if interpretation of the data did not allow to assess reliably the depth of the different horizons. Later on, it was determined that the interpretation difficulties were due to a certain anisotropy of resistivity within the Oxford clay. Seismic techniques were also applied, but their results were characterized by significant uncertainty. In consideration of the higher cost of seismic surveys, it was concluded that a situation such as the one existing at Down Ampney would be investigated best with resistivity surveys.

After the surface geophysics, a number of boreholes were drilled through the fault zone to determine its hydraulic properties and to carry out down-hole geophysical investigations. In particular an advanced procedure for cross-hole seismic tomography was tested between the four large-diameter boreholes. Despite some interpretation difficulties, the tomographic survey did show the existence of a lower velocity layer in the fault zone. This is consistent with an enhancement of porosity reasonably due to diffuse fracturing. Subsequent hydraulic testing (pulse tests) confirmed that the permeability in the fault zone was increased over the surrounding undisturbed clays between one and two orders of magnitude.

Geochemical analyses in the fault zone confirmed that the fault had a significant impact on solute transport.

In conclusion the Down Ampney investigations have demonstrated that faults can indeed affect the barrier function of Oxford clay.

**Orte Investigations**

As said before the Orte quarry is located in blue clays of Plio-Pleistocene age. At the time of the activities described in this paper the quarry was inactive, having been shut down a short time before; however the steps and the excavation faces were relatively fresh, allowing to recognize a variety of features, including some small faults, among which one with a throw of about one meter. Due to the relatively homogeneous nature of the Orte clays, to their thickness (over 300 m) and to the known fact that the fault did not cause drastic alterations of clay properties, the detection of the fault by geophysical methods was perceived from the very beginning as a real challenge. For this reason a significant effort was spent in improving the resolution of the reflection seismic technique. A variety of energy sources, of geophone configurations and of data elaboration methods were tested. For the Orte clay, the betsy (seismic gun, caliber 8 mm) turned out to be the best energy source. In addition geoelectrical soundings were carried out in the quarry area and in its proximity.

In the end the geophysical methods employed at Orte were judged to achieve a resolution capability of about one meter, which made detection of the existing fault somewhat marginal.

Later on two 90 m boreholes were drilled to intercept the fault plane. Discontinuities were actually cored in the right depth range, which makes successful interception of the fault very likely. On the other end the possibility of underground bending of the fault plane and of interception of a different unknown feature cannot be ruled out entirely.

The boreholes allowed some additional investigations, including: borehole logging, hydraulic testing, geochemical analyses on the solid phase and pore water and geotechnical testing of clay samples.
The hydraulic tests failed to reveal an increase of permeability in the vicinity of the fault, which confirms the laboratory determinations carried out a few years before. The geochemical analyses, on the other hand, have indicated that, near the fault, some migration processes must have taken place. Evidence for this is provided by the disequilibrium of uranium isotopes, observed in some samples, and by the variation of concentration of redox sensitive species in the vicinity of the fault plane (Baldi et al., 1991). The interpretation of these findings can be either that some fluid migration has taken place in the past and was later stopped by self healing of the fault or that some enhanced migration is still going on but the increase of permeability is too small to be detected by hydraulic testing.

**Narni Investigations**
The Narni study site is located on a small hill above an active clay quarry where a fault with a throw between three and four meters was known to exist. The fault was clearly visible before 1987, but then it was hidden by a slide of the quarry front. The Ismes investigations at Narni were limited to two high resolution reflection seismic lines, two resistivity profiles, five geoelectrical soundings and a structural survey of the area. The greater throw of the fault and the presence in the clay body of some lignite layers (good seismic reflectors) caused favourable expectations on the capability of geophysical methods to detect the dislocation. However, even at Narni, the measured anomalies were difficult to correlate and their interpretation remained hopelessly ambiguous.

**Soil Gas Surveys**
Exeter University contributed to the Faults in Clays project by performing soil gas surveys at Down Ampney and at Narni.

At Down Ampney, soil gas samples were collected along north-south traverses; sampling was effected at a depth of 0.5 m, every 12.5 m. Samples, analyzed for $^4$He, radon, CO$_2$ and O$_2$, revealed anomalous concentrations in two major linear zones. One, in close proximity to the fault already identified by BGS, showed high levels of $^{222}$Rn, suggesting the existence of a high permeability pathway, since the short life of the radionuclide implies fast travel times.

To explore the capability of the soil gas method and the rate of gas flow along the fault zone a gas injection was carried out. Helium and CO$_2$ were injected in an aquifer horizon, near the fault plane, at a depth in excess of 30 m. High helium levels were measured at the surface about 20 hours after the injection, confirming the permeability of the fault zone.

At Narni helium and CO$_2$ were measured on a short trial traverse, followed by the full complement of gas analyses on two longer traverses. Sampling was performed every ten meters. Radon and helium anomalies were identified and interpreted as an indication of a north-south trending fracture. The soil gas data appear to support the location and the tectonic nature of an ambiguous feature identified by Ismes with the geoelectrical survey.

Later on teams from the universities of Exeter and Rome have continued to perform soil gas surveys, such as the work in the framework of the EC project on "The refinement of soil gas analysis as a geological investigative technique" recently concluded. It is worth mentioning that within this project a site in Tuscany has been investigated with a combination of techniques, including geophysics and soil gas surveys. In this case Ismes has succeeded in locating by seismic reflection a near-surface fault plane in clay that, subsequently, was successfully used for a gas injection test (Lombardi et al., 1996).
3. CONCLUSIONS

The information reviewed in the preceding pages seems to justify a number of conclusions.

- In some cases fractured clays can show a hydraulic conductivity that is locally enhanced as much as two or maybe three orders of magnitude. At other locations, fractures do not appear to cause any noticeable water migration. Different behaviour can be explained by differences of intrinsic properties, but other factors could also cause different responses, for example relative orientation of fracture planes in respect to major stress in the rock is expected to have a significant impact on transmissivity of the discontinuities.
- Direct observational evidence for fracture permeability in clays is limited. Some information exists but is usually qualitative and poorly characterized. Part of the problem is the relative lack of in situ measurements and deep cores from clay formations. Consequently the reasons for the different hydraulic behaviour, even for apparently similar clays, are not fully understood.
- The observations described in this paper are frequently derived from sources that at the time were involved in engineering projects and had a marginal interest in the hydraulic properties of clays.
- Despite the limitations due to the uncertainty of many data, the picture of a complex and difficult-to-predict behaviour is confirmed.
- Due to the many factors controlling the physical and hydraulic properties of argillaceous sediments, it is practically impossible to predict the secondary permeability at depth without extensive site specific investigations.
- Geophysical techniques by themselves are of little use in locating discontinuities in clays and in determining their hydraulic properties.
- The most promising approach for the characterization of clay sites and the assessment of the isolation capacity of the clay layers consists in using a variety of techniques. A sensible integration of geophysics, geochemistry (including soil gas analyses), hydrogeology, borehole drilling, down-hole measurements and sample testing should allow to assess reliably the barrier capacity of the clay.

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Soil Gases as Fault Tracers in Clay Basins.  
Two Case Histories: The Bovey (UK) and Siena (I) Basins

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Abstract

The upward migration of gases generated naturally in the Earth's crust, such as helium, radon, carbon dioxide, hydrogen and methane, occurs preferentially along tectonic discontinuities, which form zones of enhanced secondary permeability. High concentrations of these tracer gases in the near-surface soil can be interpreted as a distinctive signal of gas migration to the surface and provide a detection method for gas-permeable faults. The soil-gas method may be applied to the safety assessment of the far-field geological barriers of radioactive waste repositories, where faults may provide pathways for radionuclides to pass into the biosphere. The gas permeability of faults in clay formations was assessed by studying natural tracer-gas (He, Rn and CO₂) concentrations over known faults in clay basins in the UK (Bovey Basin) and Italy (Siena Basin).

The Bovey Basin consists of at least 1000 m of Eocene and Oligocene sand, clay and lignite in a pull-apart basin associated with a major fault zone trending NW-SE across Devon. A regional soil-gas survey was conducted over 57 km² at 2 samples/km². Contoured concentration plots of the soil gases show concentric positive anomalies associated with faulting, particularly at the basin margins. Detailed traverses and surveys down to 10 m sample spacing were also performed at some localities to determine the signal over minor intra-basinal faults.

The fault bounded Siena Basin is characterised by the occurrence, below a clay cover up to 2000 m thick, of high-pressure gas domains (linked to low enthalpy geothermal reservoirs). It thus shows some similarities to an underground radioactive waste repository in clay, with gas under pressure. The basin is cut into three parts by two transverse fault systems: the Aria Line and the Grosseto-Pienza Line which are major SW-NE regional discontinuities. A regional soil-gas survey (1 sample/km²) for He, Rn and CO₂ was made throughout the northern sector of the basin. More detailed surveys were performed in an area crossed by the Arbia line, over different sized grids down to 50 m sample spacing and along traverses, to define the sensitivity of the soil-gas method for tracing faults.

The soil-gas surveys show that leakage of terrestrial gases corresponds with tectonic fracture zones, both along the margin and within the centre of the clay basins. In spite of the great thickness and plasticity of the clay, if fractured, clay does not always form an impermeable barrier for migrating gas. The localisation of gas anomalies at points along faults suggests that channelling occurs within the fault network indicating the ability of clay to be a seal along parts of the structure. The spatial association between He, Rn and CO₂ concentrations suggests that CO₂ may act as a carrier gas for the trace gases which, because of their very low concentrations, cannot advect on their own. Gas migration through clay formations seems to follow advective processes, as supported by exhalation data.
Introduction

This study is part of an EC-funded project to assess the use of soil-gas surveying as an investigative technique in the characterisation of potential sites for the disposal of radioactive waste in clay formations [1]. The primary goal was to assess to what extent clays, owing to their plasticity and low intrinsic permeability, hinder or limit gas migration.

The upward migration of gases generated naturally in the Earth's crust, such as helium, radon, carbon dioxide, hydrogen and methane, occurs preferentially along tectonic discontinuities, which form zones of enhanced secondary permeability. As reported in the literature, high concentrations of these naturally occurring tracer gases in the near-surface soil can be interpreted as a distinctive signal of gas migration to the surface. The soil-gas technique thus provides an independent fault-detection method for gas permeable faults or may enhance results when coupled with other field surveys [2]. For these reasons the soil-gas method may be applied to the safety assessment of the far-field barriers of proposed and existing radioactive waste repositories, where faults may provide pathways for radionuclides to pass into the surrounding environment. The gas permeability of faults in clay formations was assessed by studying natural tracer-gas (He, Rn and CO₂) concentrations over known faults in clay basins in the UK (Bovey Basin) and Italy (Siena Basin).

Analysis

Soil-gas samples for both the UK and Italian work were analysed by similar methods. The soil-gas samples were collected at a depth of about 0.5 m by extracting the gas from the pore space of the soil via temporarily emplaced stainless steel probes. The Siena Basin samples collected by the University of Rome were transported in small stainless steel cylinders to the laboratory and analysed by mass spectrometry (He) and gas chromatography. Those collected by the University of Exeter in both the UK and work in the Siena Basin were stored in polypropylene syringes and analysed within 24 hours. A portable infra-red analyser was used for CO₂ analysis in the Bovey Basin and gas chromatography where samples exceeded the 5% maximum of the instrument. The Rn activity was determined in the field by alpha-particle counting.

Case History 1: The Bovey Basin

The Bovey Basin (Devon, UK) is a flat area mostly at heights of 10 to 30 m above sea level, in which the rivers Bovey and Teign join to flow south-east. Their courses may at least in part be related to underlying tectonic trends and this in itself suggests that the structure of the clay basin may be related to subsurface faults. The high ground of the Dartmoor granite dominates to the north-west of Bovey Tracey. The Tertiary basin consists of at least 1000 m of Eocene and Oligocene sand, clay and lignite within a pull-apart basin associated with the Sticklepath Fault, a major tectonic feature consisting of 80 km of en-echelon faults trending NW-SE across Devon. The surrounding Palaeozoic formations show dextral offset up to almost 5 km and the post-Variscan Dartmoor granite has a 1.3 km dextral displacement along its southern margin. Devonian rocks are seen to overthrust the southern margin of the Bovey Basin, providing further evidence of the Tertiary activity of this fault.

A regional soil gas survey of He, Rn and CO₂ was conducted randomly over 57 km² at a density of 2 samples/km² [3]. Radon 220 and 222 were differentiated using the method of Morse [4]. The effects of meteorological variations were minimised by spreading out the points surveyed each day at random over the whole area. In addition the survey was first completed across the entire area at 1 sample/km², before doubling this density.
Contoured concentration plots (Figures 1 and 2) of the soil gases, together with the geology, show that the areas of anomalous gas are mostly associated with faulting, particularly at the basin margins. Although some high soil gas concentrations appear as isolated anomalies, many form contiguous features. The NW part of the surveyed area (which crosses the Sticklepath Fault Zone) has no associated He anomaly but does show high CO₂ and Rn values. Radon is expected to be high at this location due to the proximity of the Dartmoor granite, but it is also anomalous within the clay basin. In addition CO₂ forms a linear NW-SE trend in the basin that is associated with He and Rn; this is aligned with the Sticklepath Fault, suggesting that the fault might be present at depth and that fractures within the clay are allowing gas transport. Within the basin from near Bovey Tracey (822 781) to New Bridge near Chudleigh Knighton (850 765) there is an elongated zone of soil gas anomalies represented in places by one or a combination of elevated He, Rn and CO₂. The published map shows NNW-SSE and NW-SE faults in this area which may provide gas permeable pathways, or possibly the anomalies are associated with an extension of the Sticklepath Fault system.

The Twinyeo Farm site (846 760) is situated on the eastern side of the Bovey Basin within the area of economic clay working. The published geological map shows a NNW-SSE fault passing beneath Twinyeo Farm and downthrowing to the east. This fault is reported as being visible in the Newbridge workings to the north. Soil-gas samples were collected for He, CO₂ and Rn from 67 sample sites (Figures 4 & 5) and along two traverses (1 and 2). Borehole data suggests two faults, labelled A and B on Figures 4 & 5. The soil gas concentrations support the presence of faulting, most striking being Traverse 1 (Figure 3) where ΔHe is anomalous over the fault compared to adjacent background values of around 50 to 100 ppb. From 120 to 160 m ΔHe shows consecutive values of 213, 320, 352, 522 and 181 ppb. Carbon dioxide is also high, peaking at 5.57%, but perhaps equally significant are the O₂ and N₂ values at this location at 2.14% and 90.43% respectively. These could suggest that an oxidation event has depleted the soil gas atmosphere which under normal biogenic conditions would be restricted to O₂ values of about 17 to 20% and N₂ around 79%. Such an oxidation might occur in the presence of methane, but the maximum recorded was 0.025% at 167 m where the traverse intersected the field hedge. Carbon dioxide would probably be significantly higher than 6% under these conditions. Upwelling of gases from a fault therefore seems a strong possibility.

The ²²⁰Rn distribution (Figure 4) at 8460 7607 and 8448 7617 would support a NW trending fault and this agrees with the surface fault identified by the clay company, here referred to as Fault A. Helium is also anomalous at 8453 7613, but is not elsewhere along Fault A. All the soil gas anomalies are of limited linear extent suggesting the possible pipe-like nature of fault permeability to gases. The highest ²²⁰Rn (Figure 5) occurs in the Bovey Valley and may correspond to Fault B, but it may also be that there is much surface granite (uranium source) material in the valley.

A regional approach to soil gas analysis as an aid to geological interpretation of an area, can reveal information even at a 1 to 2 sample/km² density. Data collected for He, Rn and CO₂ produces patterns on contoured maps that are more than just random sets of data associated with, for example, meteorological factors and analytical error. In places, anomalous values of He and CO₂ strongly suggest an association with faulting, particularly where associated with anomalous Rn, without a corresponding high soil U or Th content. The work at Twinyeo Farm has shown the value of small-scale detailed surveys to determine the presence of faults, particularly in cases such as this where there is no surface expression of faults. Such faults may of course be missed on the regional surveys, but even on the detailed scale the combination of grid surveys followed by traverses with 10 m sample spacing may be needed to elucidate the permeable sections of faults.
Figure 1. Contoured soil-gas concentration plots ($\Delta$He in ppb, left, and $^{222}$Rn in Bq/l, right) for the Bovey Basin (white background).

Figure 2. Contoured soil-gas concentration plots ($CO_2$ in $\%$, left, and $^{220}$Rn in Bq/l, right) for the Bovey Basin (white background).
Figure 3. Traverse 1: soil-gas survey at Twinyeo Farm, Bovey Basin.

Figure 4. Soil gas $^{222}$Rn at Twinyeo Farm, Bovey Basin.
Figure 5. Soil gas $^{222}\text{Rn}$ at Twanye Farm, Bovey Basin.

Case History 2: The Siena Basin

The Siena basin was selected as a research site for soil gas analysis as it shows some similarities to an underground radioactive waste repository where gas can be under high pressure. In fact the basin is characterised by the occurrence, below a thick clay cover (up to 2000 m), of high-pressure gas domains (linked to low enthalpy geothermal reservoirs). The formation of the Neogene Siena basin followed the change from a compressional period during the Upper Tortonian to one of tensile tectonics which produced extensional structures. The basement, thrust towards the NE is believed to be present at 1 to 1.5 km depth beneath Pliocene sediments, which are mainly marine deposits of clays, sandy-clays and sands with conglomerates. The main boundary fault on the NE side of the basin lies near Rapolano Terme and has a throw in the order of 2000 m; in contrast the SW boundary is composed of a fault system, with each fault having a lower individual throw. The elongated basin is cut into three parts by two transverse fault systems: the Arbia Line and the Grosseto-Pienza Line. These structures are major SW-NE regional discontinuities and are visible on maps and satellite images (Figure 6). Two sites (1 and 2) were chosen for detailed study.

At site 1, a survey in a restricted area of about 59 km$^2$ (3-4 samples/km$^2$) was performed around the Acqua Borra geothermal spring, which discharges in the middle of the basin. Since soil moisture and meteorological factors such as rainfall affect the measured values, sampling was carried out in the summer period which is the driest and meteorologically most homogeneous season. The results are shown in Figures 6 and 7. The observed soil gas distributions confirm the presence of two fracture zones within the clay basin which conduct gas from the basement to the surface. The first zone fits well with the Arbia Line fault system whilst the second lies over a minor fault system crossing the Arbia line.
Figure 6. Location of Siena Basin (above) and contoured ΔHe soil gas concentrations at Acqua Borra (below).
Figure 7. Contoured soil-gas Rn activity (above) and CO₂ at Acqua Borra (below).
At site 2, further ENE along the Arbia line, more soil gas surveying showed that higher CO$_2$ and Rn values (Figure 8) were linearly distributed in the central sector of the investigated area. The anomalies appear to follow the Malena river including its changes of direction which are likely to have been induced by tectonics. On the contrary the major He concentrations were found on the SE side of the surveyed area though a He anomaly was found by more detailed traverse surveys close to the Malena River (Figure 9). The traverse was aligned on a NNW-SSE direction and both He and $^{222}$Rn showed an anomaly with a peak that was asymmetrical with the steep southern side corresponding closely to the position of the Malena River and the suspected position of the Arbia Fault line. The unsmoothed $\Delta$He peak at this point is 342 ppb at site A-15 and $^{222}$Rn 52 Bq/l at site A-14. Carbon dioxide is also at its highest at 7.68% and oxygen is anomalously low at 8.8%. It is therefore clear from the soil gas data that this is the location of a major permeable fracture, the Arbia Line.

Figure 8. Contoured soil gas Rn activity at Pieve di Pacina, Siena Basin.
Figure 9. Traverse from SSE (left) to NNW (right): soil gas ΔHe at Pieve di Pacina, Siena Basin.

A regional (1 sample/km²) soil-gas survey for He, CO₂, and Rn was made throughout the northernmost sector of the Siena basin to study gas migration in faulted clay when high gas pressure gradients (the geothermal reservoir) are present. The survey showed that most of the higher soil gas concentrations were located along the fault systems. Though the mean ΔHe was only 8 ppb, concentrations up to a maximum of 626 ppb suggest the occurrence of a deep origin of gas upflow. Radon values at some points are above the levels that may be supported by the low uranium content of the clays [5]. Only an advective gas upflow from the subsoil can account for the observed radon anomalies. Carbon dioxide at one point reached a maximum of 46% three orders of magnitude above the atmospheric content. Statistically derived contourlines of Rn, CO₂, and ΔHe anomalies showed similar locations, shapes and directions (Figures 10).

Figure 10. Contoured soil-gas data from the Siena Basin (Rn above, CO₂ and ΔHe next page)
Figure 10. (cont.) Contoured soil-gas data from the Siena Basin (CO₂ above and ΔHe below)
Conclusions

The main results of the gas leakage study by soil-gas surveys showed that leakage of terrestrial gases occurs in correspondence with tectonic fracture zones, both along the margin and within the centre of clay basins. This observation means that in spite of the great thickness and plasticity of the clay sequence, if fractured it does not always form an impermeable barrier for naturally migrating gas; clays can prevent gas from rising only far away from highly fractured zones. On the other hand, channelling observed within the fault network may be an indicator of the ability of clay to seal itself along certain portions of the structure.

The spatial association between Rn, He and CO₂ concentration suggests that CO₂ may act as a carrier gas for trace gases which, because of their very low concentrations, cannot advect on their own. In fact gas migration through clay formations seems to follow advective processes, a theory also supported by exhalation data.

The results from the two scales of sampling grids, performed both in the Bovey and Siena basins, show an appreciable congruence that can be observed for all the gases studied.

References


Heterogeneity of Hydraulic Conductivity of a Fault in Sedimentary Sequences at the Tono Mine, Central Japan

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Abstract

This study aims to evaluate the hydraulic conductivity of a fault zone in sedimentary sequences and to develop hydrogeological models of faults. The hydrological conditions on either side of a fault in Tertiary sedimentary sequences, known as the Tsukiyoshi Fault, were compared to make qualitative estimates of the fault zone hydraulic conductivity. The hydrological data were obtained by long-term piezometric head observation as part of the Shaft Excavation Effect Experiment, in which a shaft, 6 m in diameter and 150 m in depth, was excavated at the Tono Mine, central Japan.

Prior to shaft excavation, the heads measured in boreholes on the upthrown side of the fault were lowered by the presence of the existing mine gallery located on the same side of the fault. Heads measured on the opposite, downthrown side were not affected by the existing gallery. The transient behavior of piezometric drawdown during and after the shaft excavation was different on either side of the Tsukiyoshi Fault. On the upthrown side, the heads were drawn down simultaneously with the shaft excavation. On the downthrown side the heads were drawn down more slowly. A comparison between changes of piezometric head and results of in-situ hydraulic tests revealed no relation between the size and transient behavior of piezometric drawdown and the hydraulic conductivity of the immediately surrounding rocks.

These observations show that the fault behaves as a low conductivity zone. However, the difference in size of piezometric drawdown between the upper part and the lower part of the Toki Lignite-bearing Formation on the downthrown side implies that the hydraulic conductivity of the fault zone is heterogeneous. This conclusion agrees with the evidence which indicates that the fault clay zone in the lower part of this formation is more than two times as thick as that in the upper part. Numerical modelling is currently being undertaken to provide a more quantitative basis for these conclusions.
1. INTRODUCTION

The groundwater flow in a rock mass may be characterized by the distribution of faults and fracture zones which can act either as preferential pathways or as hydraulic barriers. Evaluation of the hydraulic properties of faults and fracture zones are of prime interest when conducting the performance assessment of geological disposal systems.

In general, the thickness and structure of the same fault will vary at each observation point. For example, the thickness of a fault zone in granitic basement was observed to vary from 65 cm to 900 cm within a distance of 500 m [1]. In particular, it is often evident that the thickness and structure of a fault vary as it cross-cuts different formations in sedimentary sequences, and consequently the hydraulic conductivity of the fault within each formation varies [2].

As a part of the hydrogeological studies, this study is being conducted to evaluate the hydraulic conductivity of a fault in sedimentary sequences and to develop hydrogeological models of faults. We discuss the heterogeneity of hydraulic conductivity of a fault based on the results of long-term piezometric head observations that have been made as part of the “Shaft Excavation Effect Experiment”, in which a shaft, 6 m in diameter and 150 m in depth, was excavated at the Tono Mine in central Japan [3]. This paper describes the long-term piezometric head observations and the conclusions about the heterogeneity of hydraulic conductivity of a fault in Tertiary sedimentary sequences.

2. GEOLOGICAL SETTING

The Tono Mine is located about 350 km south west of Tokyo. This area is the site of Japan’s most extensive uranium deposits. The geological setting of the Tono Mine is composed of Tertiary sedimentary sequences and Cretaceous granitic basement (Figure 1) [4]. Tertiary sedimentary sequences, known as the Mizunami Group, are divided into three formations on the basis of sedimentary environment. These are, in ascending order, the Toki Lignite-bearing Formation, the Akeyo Formation, and the Oidawara Formation. The Toki Lignite-bearing Formation mainly consists of conglomerate in the lower part, and interbedded sandstone and mudstone in the upper part. The Akeyo Formation consists mainly of tuffaceous sandstone. The Oidawara Formation consists mainly of siltstone and mudstone. Pliocene

Figure 1. The location and geological setting of the Tono Mine.
unconsolidated sediments of the Seto Group unconformably overlie the Mizunami Group and the granitic basement. A reverse fault is known to exist in this area. This fault, known as the Tsukiyoshi Fault, dips to the south at about 60 to 70 degrees and has a throw of about 30 m.

3. SHAFT EXCAVATION EFFECT EXPERIMENT

The Shaft Excavation Effect Experiment (SEE experiment) was started as one of PNC's geoscientific research programme in December 1989. The goal of the experimental programme is to develop a comprehensive understanding of excavation effects on the mechanical and hydraulic properties of the surrounding rock masses. In this experiment, a new shaft, 6 m in diameter and 150 m in depth, known as "No. 2 Shaft", was constructed by drill-and-blast method in the Tertiary sedimentary sequences at the Tono Mine. Figure 2 shows a schematic view of the No. 2 shaft and its excavation schedule. The experimental programme includes hydraulic conductivity measurements, piezometric head monitoring, rock displacement measurements, seismic tomography, and numerical simulations. Boreholes for these experiments were drilled around the shaft and monitoring systems were installed in these boreholes to evaluate the influence of the shaft excavation [3].

4. IN-SITU HYDRAULIC CONDUCTIVITY MEASUREMENT

Hydraulic conductivities were determined by the PNC low pressure Lugeon test system and the PNC low-water-pressure controlled hydraulic test system. These systems are able to measure hydraulic conductivities from $10^{-3}$ to $10^{-7}$ cm/sec and from $10^4$ to $10^8$ cm/sec, respectively [5]. Schematic diagrams of these system are shown in Figure 3. The hydraulic tests were performed at a total of 106 measurement intervals in ten boreholes around the No. 2 shaft. Hydraulic conductivities of sedimentary rocks and granitic basement, obtained by in-situ hydraulic tests, range from $10^{-3}$ to $10^{-4}$ cm/sec and from $10^4$ to $10^9$ cm/sec, respectively.
5. LONG-TERM PIEZOMETRIC HEAD OBSERVATION

MP systems (Multiple Piezometer system [6]) were installed in each borehole to observe the transient behavior of piezometric head caused by the shaft excavation [7]. The depth of these boreholes is about 200 m. Figure 4 shows the location of observation boreholes. MP system can take measurement of piezometric head and groundwater sampling from multiple levels within a borehole. In this study, piezometric heads have been observed at a total of 251 monitoring intervals in boreholes with MP systems. Observations began before the shaft excavation and has continued until now for over 2,000 days.

![Figure 4. Location map of boreholes for long-term piezometric head observation.](image)

6. DISCUSSION

6.1 Interpretation of the profiles and transient behavior of piezometric heads

Figure 5 shows piezometric head profiles before the shaft excavation. The Tsukiyoshi Fault exists between boreholes TH-4 and TH-6. The piezometric heads in boreholes TH-4 and TH-7 on the upthrown side are drawn down by the existing gallery, while the piezometric head profile of borehole TH-6 on the downthrown side is almost hydrostatic. These piezometric heads are considered to be in equilibrium as the existing gallery was excavated about twenty years ago. Steady-state numerical simulation confirms this hypothesis.
Figure 5. Piezometric head profiles on either side of the Tsukiyoshi Fault before the shaft excavation.

The piezometric head data of boreholes TH-2 and TH-5 were used to compare the transient behavior of piezometric heads on either side of the Tsukiyoshi Fault during and after the shaft excavation. Borehole TH-2 is 30 m away from the No. 2 shaft and is located on the upthrown side. Borehole TH-5 is 40 m away from the No. 2 shaft and intersects the fault at 70 m in depth under the surface. The distance between boreholes TH-2 and TH-5 is 30 m. The cross-section between boreholes TH-2 and TH-5 is shown in Figure 6. The transient behavior of piezometric heads in boreholes TH-2 and TH-5 are shown in Figure 7.

The piezometric drawdown in the upthrown side accorded with the shaft excavation schedule and almost attained equilibrium on completion of the shaft excavation. However, piezometric drawdown by the shaft excavation was not observed in the lower part of the Toki Lignite-bearing Formation of the upthrown side, because the piezometric heads were already drawn down to the level of the existing gallery. On the other hand, the piezometric heads on the downthrown side have been more gradually drawn down. The pattern of piezometric drawdown seems to be
unrelated to lithology, except for a conglomerate layer which is the boundary between the upper part and the lower part of the Toki Lignite-bearing Formation. These observations imply that the Tsukiyoshi Fault has low conductivity.

Figure 6. Cross-section between boreholes TH-2 and TH-5.
6.2 Heterogeneity of the fault zone hydraulic conductivity

The average of piezometric drawdown in each formation of Tertiary sedimentary sequences is shown in Table 1. On the upthrown side, the averages piezometric drawdown in the Akeyo Formation and the upper part of the Toki Lignite-bearing Formation are about 20 m, while that in the lower part of the Toki Lignite bearing Formation is about 16 m. This difference is considered to be the influence of the existing gallery. The difference of average piezometric drawdown between the upper part and the lower part of the Toki Lignite-bearing Formation is also recognized on the downthrown side. In this case, as the piezometric head profile is hydrostatic on the downthrown side before the shaft excavation, the difference of the average piezometric drawdown is not considered to be the influence of the existing gallery. The distributions of piezometric drawdown and hydraulic conductivities in borehole TH-5 are shown in Figure 8. This figure reveals that piezometric drawdown in the high conductivity zones is not necessarily big. These results imply that the hydraulic conductivity of the fault zone in the lower part of the Toki Lignite-bearing Formation is lower than that in the upper part of this formation.
Figure 9 shows two sketch diagrams of the Tsukiyoshi Fault in the galleries. The diagram (A) is the fault zone at EL. 190 m in the No. 2 measurement drift of the No. 2 shaft (see Figure 2). The geology of this part is tuffaceous sandstone (downthrown side) and conglomerate (upthrown side) of the upper part of the Toki Lignite-bearing Formation. The fault zone consists of two clay layers and a unconsolidated fine sand layer. The clay layers are 2 to 3 cm thick, and the unconsolidated fine sand layer is 10 to 20 cm thick.

The diagram (B) is the fault zone at EL. 160 m in the north extension drift (see Figure 1). The geology of this part is coarse sandstone (downthrown side) and conglomerate (upthrown side) of the lower part of the Toki Lignite-bearing Formation. The fault zone consists of a 10 to 20 cm thick clay layer. The difference of thickness and structure of the fault zone in the upper and lower part of the Toki Lignite-bearing Formation support the idea that the relative hydraulic conductivity of the Tsukiyoshi Fault in the lower part of this formation is lower than that in the upper part of this formation.

### Table 1. The average piezometric drawdown in each formation.

<table>
<thead>
<tr>
<th>Formation</th>
<th>Upthrown side</th>
<th>Downthrown side</th>
</tr>
</thead>
<tbody>
<tr>
<td>Akiyo Formation</td>
<td>20.8 m</td>
<td></td>
</tr>
<tr>
<td>Toki Lignite-bearing (Upper)</td>
<td>22.2 m</td>
<td>17.3 m</td>
</tr>
<tr>
<td>Toki Lignite-bearing (Lower)</td>
<td>16.6 m</td>
<td>10.6 m</td>
</tr>
</tbody>
</table>

### Figure 8. The distribution of piezometric drawdown and hydraulic conductivity in borehole TH-5.

Figure 9. Sketch diagrams of the fault zones in the No. 2 measurement drift (A) and the north extension drift (B).
Moreover, as the piezometric drawdown in conglomerate layers nearby EL. 160 m, the boundary between the upper part and the lower part of the Toki Lignite-bearing Formation in the downthrown side, accorded with the shaft excavation schedule as well as that in the upthrown side as shown in Figure 7, hydraulic pathways could be formed within the fault zone in these layers. This also enhance the applicability of hypothesis in this study.

7. SUMMARY AND CONCLUSIONS

Monitoring of piezometric heads has been conducted around the No. 2 shaft to evaluate the hydrological effect of shaft excavation in Tertiary sedimentary sequences. In this experiment, piezometric heads have been observed at a total of 251 monitoring intervals in boreholes with MP systems. Observation began before the shaft excavation and have continued until now for over 2,000 days. A reverse fault is known to exist in this area. This fault, known as the Tsukiyoshi Fault, dips to the south at about 60-70 degrees and has a throw of about 30 m. These observations revealed that the hydrological conditions on either side of the Tsukiyoshi Fault were different as follows;

(A) Before the shaft excavation, piezometric drawdown caused by the existing gallery was observed on the upthrown side, while on the downthrown side the piezometric head profile was hydrostatic.

(B) The transient behavior of the piezometric head on the upthrown side accorded with the shaft excavation schedule, while piezometric heads on the downthrown side were more gradually drawn down.

These observations imply that the Tsukiyoshi Fault is a low conductivity fault.

The average piezometric drawdown in the upper and lower parts of the Toki Lignite-bearing Formation on the downthrown side are different, though piezometric heads on the downthrown side are not influenced by the existing gallery. As the piezometric drawdown in the high conductivity zones is not necessarily big, this result implies that the hydraulic conductivity of the fault zone in the lower part of the Toki Lignite-bearing Formation is lower than that in the upper part of this formation. This conclusion agrees with the evidence which indicates that the fault clay zone in the lower part of the Toki Lignite-bearing Formation is more than two times as thick as that in the upper part of this formation. Moreover, as the piezometric drawdown in the conglomerate layer on the boundary between the upper and lower parts of the Toki Lignite-bearing Formation on the downthrown side, accorded with the shaft excavation schedule, as well as that on the upthrown side, hydraulic pathways could be partially formed in the fault zone. Numerical simulation of the area is currently being carried out in order to make a more quantitative evaluation of these conclusions.

Consequently, hydraulic conductivity of a fault should be evaluated at each part of the same fault for the modelling of a fault, particularly in sedimentary sequences.
8. ACKNOWLEDGEMENTS

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9. REFERENCE


Hydrogeology of a Fractured Shale (Opalinus Clay): Implications for radionuclide migration

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Abstract

As part of the Swiss high-level radioactive waste disposal programme, Nagra is currently investigating the Opalinus Clay, a 95 - 120 m thick Middle Jurassic shale (claystone) formation, with potential siting areas in Northern Switzerland. Observations in clay pits, and the results of a German research programme focusing on hazardous waste disposal aspects, have demonstrated that the near-surface Opalinus Clay is rather permeable. Between depths of 10 to 30 m, the permeability decreases drastically and saline waters have been sampled in a borehole at 50 m depth. Hydraulic tests in deeper boreholes, where the Opalinus Clay was encountered at depths between 300 and 1100 m, yielded very low hydraulic conductivities (< $10^{-12}$ m/s) even though joints and faults were included in some of the test intervals. These measurements are consistent with hydrogeological information from Opalinus Clay sections of six railway tunnels and four motorway tunnels in the Folded Jura of Northern Switzerland. Despite the complex tectonics of the Folded Jura, comprising numerous reverse (thrust) and normal faults in a water-saturated environment, only two indications of humid patches (one in a fault containing calcite veins, one of unknown origin) have been reported from a total of 6400 m of tunnel sections in the Opalinus Clay. The only measurable water inflow reported was in connection with an intercalated calcareous sandstone layer, which is only developed in the north-western edge of Switzerland. The hydrogeological dataset presented in this paper - part of which is for a worst-case type geological environment - suggests that radionuclide transport through faults and joints is probably not a critical issue for assessing the suitability of Opalinus Clay as a host rock for a deep geological repository for nuclear waste.
1. Introduction

As part of the Swiss high-level radioactive waste disposal programme, Nagra is currently investigating the Opalinus Clay, a shale (claystone) formation of Middle Jurassic age (Lower Dogger, mainly Aalenian). The formation - named after the ammonite *Leioceras opalinum* - consists of well-consolidated, dark grey, micaceous shales, partly with thin sandy lenses, limestone concretions or siderite nodules. The clay mineral content varies between 40-80 wt% (18-36% illite, 6-12% chlorite, 10-20% kaolinite, 6-12% illite/smectite mixedlayers in a ratio 70/30); other minerals are quartz (18%), carbonates (5-20%), feldspars (1%), pyrite (1%) and organic carbon (0.7%). The water content is in the range of 5-10%.

First scoping studies of the Opalinus Clay started at the end of the eighties with a desk study (Nagra 1988). This included results from earlier Nagra boreholes focusing on crystalline host rocks in Northern Switzerland, where the sedimentary cover of the basement, including the Opalinus Clay, was studied in detail (see section 2.2). On the basis of a reflection seismic survey, a potential siting region for a final repository for long-lived intermediate level and high-level radioactive waste has been selected in Northeastern Switzerland, where the host rock formation occurs as a 95-120 m thick subhorizontal layer in a relatively undisturbed tectonic environment (Nagra 1994, Naef et al. 1995).

The Opalinus Clay is also being studied in a service tunnel of the Mt. Terri motorway tunnel that cuts through an anticlinal structure in the Jura mountains of Northwestern Switzerland. These investigations are being performed within the framework of an international research project focusing on radioactive waste disposal issues in clays (International Mt. Terri Project). The project started with the excavation of niches and a first drilling campaign in early 1996, and to date, has yielded only preliminary, unpublished data. The main aims of the programme are to test and improve techniques for hydrogeological, hydrochemical and geotechnical investigations in an argillaceous formation and to characterise the Opalinus Clay. Some of the experiments are designed to improve the understanding of flow and transport processes, e.g. to identify preferential flow paths by tracer and resin injection, fluid logging and hydraulic testing.

2. Hydraulic Properties

2.1 Near-surface investigations

Observations in clay pits of Northern Switzerland and Southern Germany (Mazurek et al. 1996, unpublished data from the author) show evidence of a well developed near-surface groundwater circulation system in the Opalinus Clay. In the uppermost 10 to 15 m, joint systems and faults show oxidation rims with thicknesses of several centimetres and gypsum is found on fracture surfaces below the oxidation front.

In the framework of a German research project focusing on hazardous waste disposal aspects (Hekel 1994), a large number of different types of hydraulic tests in 47 shallow boreholes, accompanied by core analyses, have been performed in the Opalinus Clay of the Swabian Alb (Southern Germany). The results display a consistent picture with an uppermost 10 to 30 m thick layer with rather high hydraulic conductivities in the range of $10^{-7}$ - $10^{-4}$ m/s, containing horizontal fractures caused by stress release due to erosion of the overlying strata. Between 10 and 30 m depth the permeability decreases drastically by several orders of magnitude (Fig. 1) and stagnant saline waters have been encountered at 50 m depth in one borehole. The thickness of the permeable domain
is closely related to the geomorphological development and the topographic relief at particular sites, i.e. thicker zones of chemical and mechanical disintegration (up to 30 m) are developed below old valleys and topographic ridges, thinner zones of groundwater circulations are found below geologically young valley systems.

Figure 1. Typical hydraulic conductivity (k) profiles through the near-surface Opalinus Clay and inverse correlation of hydraulic conductivity with chloride content of porewater (data from leaching experiments), after Hekel (1994). - V_p-V_v, weathering zonation (degree of weathering) after Einsele (1993). Boreholes KB5 and KB7 from Mössingen site, Swabian Alb, Germany.

2.2 Hydraulic testing in deep boreholes

Hydraulic tests in the Opalinus Clay and adjacent layers from three Nagra boreholes and four hydrocarbon exploration wells in Northern Switzerland have been analyzed in detail (Johns et al. 1994). The tests were open- and cased-hole drill-stem tests for the hydrocarbon wells and open hole single and double packer tests for the Nagra boreholes. Although some of the boreholes penetrated joints and faults, the tests in the Opalinus Clay yielded very low hydraulic conductivities in the range of $10^{-14}$ - $10^{-12}$ m/s.
Figure 2. Example of hydrogeological mapping of exploration tunnels 3 and 4 for the Belchen motorway tunnel: Section cross-cutting Keuper marls, Liassic limestones and a small part of the Opalinus Clay. From Fröhlicher (1966). Numerous detailed indications of humid patches and water inflow outside the Opalinus Clay as for example 1: ‘etwas feucht’ (slightly humid) or 2: ‘tropft ab Decke −0.1 l/min, 17.10.64’ (dripping from ceiling −0.1 l/min). Only one humid patch (in connection with a fault containing calcite veins, not shown in this figure) has been reported from the 4 exploration tunnels, each crosscutting 500 m of Opalinus Clay.

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2.3 Observations in tunnels

Hydrogeological information from Opalinus Clay sections of six railway tunnels and four motorway tunnels in the Folded Jura of Northern Switzerland have been compiled by Gautschi (1994). The tunnels were constructed between 1860 and 1991. Most data originate from the exploration/excavation phase and the data quality ranges from general descriptions of major water inflows to very detailed mapping of minor seepages and humid patches (example in Fig. 2). The overburden in the Opalinus Clay sections varies between 100 and 800 m. Despite the complex tectonics of the Folded Jura, comprising numerous reverse (thrust) and normal faults in a water-saturated environment, only two indications of humid patches (one in a fault containing calcite veins, one of unknown origin) have been reported from a total of 6400 m of tunnel sections in the Opalinus Clay. The only measurable water inflow reported was in connection with an intercalated calcareous sandstone layer, which is only developed in the north-western edge of Switzerland.

2.4 Evidence of non-hydraulic effects

Evidence of non-hydraulic (= non-Darcian or non-ideal) effects are known from ongoing tests from the International Mt. Terri Project, where the different behaviour of the water table in boreholes containing different types of drilling fluids (deionized water, KCl brine, bentonite mud) can only be explained by chemico-osmotic effects (P. Bossart, pers. comm.). Since some of the ongoing and planned experiments are specially focused on the issue of non-ideality, future results will give a more profound basis for the discussion of these processes.

Other evidence of non-hydraulic effects in the Opalinus Clay is derived from reported long-term pressure evolution in shallow boreholes in the Homburg valley in Northern Switzerland (Horsemann et al. 1991). However, this interpretation is not unambiguous, because alternative simple explanations for the pressure response, such as the presence of air in the test intervals, cannot be totally excluded.

Non-hydraulic effects were not explicitly observed in any of the hydraulic tests in the deep boreholes of Northern Switzerland (Johns et al. 1994). All pressure responses could be easily explained by simple Darcy flow using simple flow models. However, low anomalous heads in the Opalinus Clay of one borehole (Schafisheim) may exist, but all these tests have a very large uncertainty due to their short duration. More and better quality data would be needed to identify non-ideal processes unequivocally.

3. Geochemistry

A data set comprising chemical analyses and environmental isotope measurements ($\delta^1$H, $\delta^{18}$O, Tritium) of more than 60 water samples from shallow boreholes and open clay pits in the Opalinus Clay as well as chloride leaching data from rock samples are published by Hekel (1994). Porewaters from greater depth recovered by squeezing of rock samples in a specially constructed compression rig (Entwistle & Reeder 1993), are reported by Gautschi et al. (1993). The latter samples are from the Mt. Terri Tunnel, where ongoing geochemical investigations will yield additional chemical and isotopic data in the near future.
The chemistry of the near-surface waters displays a marked depth zonation. The uppermost waters contain tritium and are of Ca-Mg-SO$_4$-HCO$_3$ type. They are part of a shallow groundwater circulation system with relatively short underground residence times (< 40 years). Their chemistry strongly reflects the geochemical weathering of the Opalinus Clay, i.e. carbonate dissolution, pyrite and siderite oxidation causing first iron hydroxide and gypsum precipitation followed by subsequent gypsum dissolution during the evolution of the oxidation front. With increasing underground residence time ion exchange processes become more important and the near-surface waters evolve gradually into tritium poor Na-HCO$_3$-(SO$_4$-Cl) waters. Between 10 and 30 m depth a marked increase in salinity (Fig. 1) clearly shows the transition into the zone of stagnant Na-Cl porewaters where diffusion is the dominant migration mechanism. The saline porewaters have salinities (total dissolved solids) up to 20 g/l, $\delta^2$H and $\delta^{18}$O values heavier than recent recharge and chloride/bromide ratios close to seawater (Gautschi et al. 1993). The chemical and isotopic data suggest that the suite of samples from high to low salinities represents a continuum produced by progressive dilution – and some water/rock interaction – of a connate formation water by meteoric recharge of various ages.

4. Significance of the hydrogeological characteristics of the Opalinus Clay for radionuclide transport

In fractured rock it is normally assumed that advective-dispersive flow occurs predominantly in the discontinuities present. When assessing solute (radionuclide) transport, it is important to also consider the potential of diffusion from such advective flow paths into the connected porosity of the surrounding rock matrix (e.g. Grisak & Pickens 1980; Neretnieks 1980). Such 'matrix diffusion' not only increases the area of mineral surfaces which can sorb radionuclides, but also provides a volume of dead space which can significantly retard even non-sorbing radionuclides.

Potential features that could decrease the effectiveness of radionuclide retardation are: the existence of preferential pathways within the discontinuities, i.e. flow channels (=> volume of rock in contact with flowing groundwater is reduced), the sealing of fracture surfaces by non-sorbing, non-porous mineral coatings (=> radionuclides have no access to the rock matrix) and high fracture transmissivities (=> radionuclides have not enough time to diffuse into the rock matrix).

The observations made on near-surface Opalinus Clay clearly demonstrate that fracture flow and matrix diffusion are dominant processes at shallow depths (cf. Mazurek et al. 1996). The occurrence of springs in clay pits and the high hydraulic conductivities measured in shallow boreholes may raise doubt about the suitability of Opalinus Clay as host rock for radioactive waste (at least in the opinion of the public). However, the extensive data set discussed in this paper indicates that such high permeability is a surface related decompression and weathering effect. The large number of faults and joints that have been penetrated by deep boreholes and by 6400 m of tunnels with only two indications of minor seepages are a good statistical basis for the conclusion that significant advective flow through fractures occurs rarely in deep Opalinus Clay. There have been no indications of narrow flow channels like those described from crystalline rocks (e.g. Thury et al. 1994), while calcite in a few of the fractures was the only hint for fracture coatings that have to be investigated in more detail in the future. Indeed, one may ask the question whether, at least for a large part of the Opalinus Clay, fracture flow is negligible and diffusion through the bulk rock mass alone will be the dominant process for radionuclide transport. In this case, it can be clearly shown, that even a few meters of good rock would provide an extremely efficient transport barrier for all radionuclides.
To answer this type of question a series of experiments is planned to be performed in the framework of the International Mont Terri Project.

5. Conclusions

The Opalinus Clay is a well-consolidated claystone/shale formation containing joints and faults. The near-surface Opalinus Clay (uppermost 10 - 30 m) is rather permeable \((k = 10^{-7} - 10^{4} \text{ m/s})\) due to chemical/mechanical weathering and erosional decompression. At greater depth (several hundreds of metres), the hydraulic conductivity is in the range of \(10^{14} - 10^{12} \text{ m/s}\). The measurements from deep boreholes in Northern Switzerland are consistent with hydrogeological information from Opalinus Clay sections of ten tunnels in the Folded Jura where, despite the complex tectonics of the Folded Jura, only two indications of humid patches have been reported from a total of 6400 m of tunnel sections in the Opalinus Clay. There are also indications of chemico-osmotic effects (Mt. Terri, Homburger Valley), which are consistent with very low permeabilities of the formation.

Porewater and groundwater in permeable fractures evolve with depth from Ca-Mg-SO\(_4\)-HCO\(_3\), type to Na-HCO\(_3\)-SO\(_4\), to Na-Cl-SO\(_4\), type (stagnant formation water), with a pronounced salinity increase between 10 and 30 m depth.

The hydrogeological dataset presented - part of which (tunnels in the Folded Jura) is for a worst-case type geological environment - suggests that radionuclide transport through faults and joints is probably not a critical issue for assessing the suitability of Opalinus Clay as a host rock for a deep geological repository for nuclear waste.

Acknowledgements

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References


Investigating Faults and Fractures in Argillaceous Toarcian Formation at the IPSN Tournemire Research Site

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Abstract

The Institute of Protection and Nuclear Safety (IPSN) has developed in situ research programmes concerning the confining properties of argillaceous formations at the TOURNEMIRE tunnel site (Aveyron). Investigations made on this type of formation of very low permeability conclude that transfers, if they exist, should be of very minor importance. In this case, it is necessary to take into account the existence of structures (faults or fractures) and check their actual role in possible transfers. The purpose of this paper is to describe the nature of these structures and to discuss their possible significance regarding possible past and present fluid circulations. Fracturation analysis of the Domerian and Toarcian argillaceous formation performed by means of fourteen boreholes reveals the existence of fault planes, which constitute the major paleocirculation network within the Tournemire massif. Fracturation from cores shows the existence of planes with different types of fillings (mainly calcite) of various sizes. This fracturation seems perfectly sealed and impervious; nevertheless, the making of boreholes close to the upper aquifer in a disturbed zone might create the possibility of fluid production some months after the drilling work. In situ permeability pulse tests located in fractured zones indicate local permeability values slightly higher than the “global matrix permeability” (from $10^{13}$ to $10^{14}$ m/s). Texture, mineralogy, chemical and isotopic contents of the secondary carbonates from the fractured zones have been analysed in order to determine the origins, ages and chemistry of the initial circulations. Four types of filling paragenesis have been identified: calcite alone, calcite and frambooidal pyrite, calcite and cubic pyrite, and calcite and barite. The morphology of calcite crystals is very variable, even within a same fracture. The “stratigraphy” influence on fracture filling chemistry through water-matrix interactions seems strong. Several indices point out slow water circulations. Uranium distribution cartographic and high iron contents in the calcite fillings suggest circulation of reducing fluids within the fractures. Profiles of $^{14}$C could be explained by thermal effects, but corresponding calculated temperature gradients are abnormally high: both different isotopic contents and different temperatures over time have to be envisaged for the percolation fluids. This hypothesis make it possible to consider the coexistence of two or three “groups” of fillings. The interpretation of the $^{13}$C profiles is more complicated and could impose hypotheses on both upwards and downwards fluid circulations. This ambiguity relative to the direction of the fluid flow generated by these carbonated deposits is not resolved by using $^{14}$C dating and U/Th family disequilibria. All the secondary calcites are $^{13}$C free and $^{235}$U/$^{238}$U often close to unity.
INTRODUCTION

In order to be able to perform correct waste disposal assessments, the Institute of Protection and Nuclear Safety (IPSN) has developed in situ research programmes addressing the confining properties of geological formations. For that purpose, IPSN has selected at the TOURNEMIRE site (Aveyron, France) a clay formation constituted by a 250 m thick indurated Toarcian and Domerian claystone layer, overlain by 270 meters of Bajo-Bathonian limestones. Investigations made on this type of formation of very low permeability and very small porosity lead us to conclude that transfers, if they exist, should be of very minor importance and excessively difficult to detect. However, despite these statements, another point which must be taken into account is the existence of structures (faults or fractures), of which it is necessary to check the present day role with respect possible transfers, between the aquifers above and below the Toarcian and Domerian argilites needs to be ascertained. The purpose of this paper is to describe the nature of these structures, and to discuss their possible significance regarding possible past and present fluid circulation.

STRUCTURAL CONTEXT OF THE TOURNEMIRE SITE

Knowledge of this kind of data has enable to precise the tectonic context of the site, its passed evolution, and if possible relationships between observed structures and hydraulic circulation throughout the sedimentary series to be determine.

Tectonic and Structural Data at the Regional Scale

The Causses basin (south of the French Massif Central) is a wide synclinorium structure with a N – S axis. On a kilometric scale, the model can be considered monoclinal, with the presence of faults of pluri-kilometric dimension and sporadic plio-quaternary volcanism. The Larzac area is characterised by a nearly E-W trending overall structural context corresponding to large regional accidents which affecting all the series of the Permo-Jurassique basin[5]. This structuration is cut by a regional NE-SW faults system and a faults network near to N-S.

The site of the Tournemire tunnel [2] is located in this context of sub-horizontal Jurassic sedimentary rocks which determine, from a large scale lithological standpoint, a “three-layer model”: a lower layer, made of a limestone and dolomite (Hettangian, Sinemurian and Carixian) layer, an intermediary layer of clayey lithology (Domerian, Toarcian and lower Aalenian) in which the tunnel is located, and an upper layer constituted of limestone and dolomite series (upper Aalenian, Bajoctian and Bathonian). Cenozoic and post-Cenozoic tectonics have affected the Jurassic formation [2]. The main accidents in the Millau area (frequently organised in bundles) are essentially E-W in direction. The structural model of the Tournemire area corresponds to a large regional E-W trending block and delimited by two major accidents of the nearly E-W system with a south vergence : to the north, the Cernon fault (the most important morpho-structural element of the Tournemire sector) linked to the Hospitalet du Larzac bundle which effect a contact the lower Jurassic. and middle Jurassic , and to the south, the St-Jean d'Alcapies fault. A third accident, associated with the major NE-SW structural scheme of the area, cuts across this large block : the Tournemire fault, the extension of which is limited by the Cernon accident to the north and extends by few kilometres beyond the St-Jean d’Alcapies accident to the south. In any case on a more detailed scale, the nearly N-S trending fault network concerns the whole volume of the large E-W block. At the surface, karstic events are well aligned following faults and joint passageways.

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Mesosstructures: Small Faults and Joints on the Scale of the Massif

Mesosstructure studies have dealt with a five km² sector (cf. figure 1) on the Causse formation and slopes surrounding the site [1]. Fracturation appears essentially sub-vertical, with three plane families: a predominantly NNW-SSE direction, and mean NE-SW and nearly E-W directions. Apart from these main accidents of the fault type, which delimit a sector subjected to weak tectonic effects around the tunnel, only joint passages or small faults have been detected. These structures cause little disturbance within the argilite mass. The main jointing, orthogonal to the stratification, is oriented N170. It is observable at all scales, and is expressed through joints and microfissuration in the limestone facies. This fissuration is not visible, at the outcrop scale, in the clays. Other directions are less frequent, and can be locally subordinated to the major accidents (the Cernon fault...) or to the morphology.

The course of the passageways has been charted through the argilites. Very few faults have been identified in the argilites because of their mechanical behaviour and above all because of the very small number of outcrops. A strike-slip fault in an outcrop north of the Tournemire village (cf. figure 1), has been observed with lateral displacement of about one meter in an argillaceous bank with nodules, without any deformation of the wall, but with joints filled in with calcite. The joint passage transecting « les Fournials » (cf. figure 1) crosses the argilites at an observable outcrop: it is formed, at the top of the nodulous argilites, of a bundle of wavy joints of five to height meters thick. Joints are slightly open, or sealed with calcite, and the argilites are oxidised on one to two cm on both sides. This alteration, probably recent, is made possible only when argilites are in contact with the open air; it reveals structures which could have not been seen during underground work and are probably of a little importance regarding possible flows through this argillaceous facies.

Possible Connection between the Structural Context and Underground Circulations

The detailed study of this fracturation (microtectonic analyses of the fault planes observed at the surface and in the cores) has made possible, as a first approach, a correlation between the kinematics analyses of faults and permeability on a large scale [1]. The different states of stress observed indicate a polyphasic evolution of the area. The ages of these tectonic events cannot be determine (lack of post-Jurassic series and syn-sedimentary deformations) but they can be deduced by comparison with other areas where such events are well known and dated. Thus different kinematics analysed on the fault planes are compatible with different states of stress, that is to say:

- a predominant **NNW-SSE trending compression**, seen in almost of the sites which can be assigned to the Pyrenean compression and responsible, in the Tournemire area for the re-activation of large accidents (nearly E-W) of the basement. This state of stress is characterised by a highly brittle strain of the Permo-Jurassic domain and a localised soft deformation. It has created a high density sub-vertical fracturation, with a direction close to the main axis \( \sigma_1 \), with plane apertures in the direction of the second horizontal axis (\( \sigma_2 \) or \( \sigma_3 \)) because of the extensive nature of this later. A majority of these permeable discontinuities, created by the fracture apertures, have been filled in with calcite from dissolution zones under the effect of tectonic stress. This state of stress is possibly the origin of a paleo-permeability following this N-S direction;

- an **E-W trending extension**, which can be related to the extensive Oligocene tectonics. Nevertheless, it is not very significant in the area and seems to be limited to a weak reactivation of the large accidents and to the localised formation of a few fault planes within the Permo-Jurassic cover. Microstructures associated with this state of stress correspond to the formation of mechanical striation: this tectonic regime in extension has enhanced the accentuation of the permeability along the N-S network;
- and a probable E-W trending compression. The associated deformation style does not seem intense within the area and does not seem to have modified the permeability to any real degree.

Present hydrogeological circulations in the Tournemire area are conditioned by the tectonic evolution and the state of stress type it has been affected by. There are several indications of an underground circulation within the Tournemire block : network of caves, oxidised traces on the planes, the presence of wet zones. Circulation within this « three-layer model » rock mass occurs essentially in the two dolomite/limestone formations, following the nearly N-S fracture network. In contrast, in the intermediate formation any circulation is highly improbable.

TUNNEL SCALE : STRUCTURAL DATA FROM BOREHOLES

Means and Methods

Boreholes have been drilled from the tunnel in order to determine the characteristics of the toarcian formation studied : its petrographic, mineralogical and geochemical nature, but also the geological history of the site, considering tectonic and structural aspects relative to the hydrogeological aspects. These data can be expected to yield a better understanding of the paleocirculations which have occurred within the massif through fractures, for instance. They are compared with the independent results obtained from laboratory analyses conducted to characterise the possible transfer through the Tournemire argilite [1, 2].

Fourteen boreholes have been drilled, in two sets for the purpose of obtaining a three dimensional view of the formation and of the fracturation which can be found around the tunnel : 6 boreholes along the tunnel axis (5 vertical downward : DC, CD, DI, DM, DS, and one vertical upward CA), and then 8 boreholes located in a plane perpendicular to the tunnel axis (7 boreholes situated in the upper Toarcian, ID0 vertical upward, ID90 and ID270 horizontal, ID45, ID135, ID225 ID315 at 45° and one vertical downward to the upper Domerian ID180).

Detailed structural analyses have been made on cores : a description of the structure types and of the observed sealing material. Fracturation seen on the cores is sketched by means of an original technique using scanning core imagery (360°), enabling the digitisation of the developed core images, and making possible the use of a specific software design to deal with analysis of faults and other structure [6]. Fracture orientation has been made possible by taking into account the general orientation of the stratigraphy of the formation.

Fracture and fissure families have been analysed considering the relative scale of the planes : major planes the characteristics of which indicate a good continuity throughout the rock mass (C1 category), secondary planes relatively to this first category planes (C2 category), and small planes and microfissures of limited local extension (C3 category).

Information obtained from cores is completed by a diagraphy campaign in the boreholes [3] : calliper to verify the wall behaviour of the wells, natural radioactivity, neutron porosity in order to get a qualitative evaluation of the porosities characteristics of the formations studied, and video imagery to examine de visu the behaviour of the borehole walls, especially at the levels where the presence of tectonic elements which have affected the rock mass have been detected.

Fracturation Characteristics from Cores and into Boreholes

As was observed at the surface at the same stratigraphic levels, fracturing seen from the cores, can be described into two kinds of planes : planes filled in with calcite, with thickness varying from few micrometers to several centimetres, and planes without any material.
In boreholes CD and CA (cf. figure 3), fracturation is localised in only certain sectors (it corresponds to plane families observed at the surface) [2, 8]. The distribution of this fracturation from the core can be summarily divided into seven major zones (Zone 1 to 7), where one can find open or closed fissures, sealed with well-crystallised calcite or very thin calcite, fractures of normal type, strike-slip fractures or inverse in some cases. To this general zoning, one can add some horizontal and quite thin isolated fissures, (less than a millimetre thick) and which correspond to bank-on-bank slips.

The 8 radiating boreholes area (drilled along a plane perpendicular to the tunnel axis) has revealed the presence of more important structures [2, 7]. The zone explored appears to be segmented into two compartments which are structurally different : on the west side of the tunnel, a relatively fractured compartment, and on the east side, a non fractured one (cf. figure 4). The boundary orientation between these two compartments fits with a fracturation passage in which the main nearly N-S fracturation is the most dominant orientation. Several planes crossed by the drill holes correspond to this plane family.

Borehole ID180 (vertical downwards) is located at the boundary of the fractured zone. Fracturation is concentrated by some sector all along the borehole. Five main sectors have been identified, exhibiting fractures and microfissures with thin fillings of calcite, slipping bank-on-bank-and-planes highly inclined planes, planes with well striated calcite filling. Borehole ID0 (vertical upwards) crosses a set of fractures (with calcite fillings) and some friction gouge or breccia zones. Here is a highly disturbed zone, where several plane families coexist (kinematics of normal, inverse and strike-slip faults).

Borehole ID315, also penetrating fractured zone, exhibits all along the cores a strong fracturation with the presence of fault zones, fractures, bank-on-bank slips and microfissuration. Boreholes ID225 and ID270, also located in the fractured compartment, present a strong fracturation comprised of faults and sectors with high a density of fractures and microfissures. The calcite of the fillings is strongly striated, the kinematics is of normal type, inverse or strike-slip. Boreholes ID45, ID90 and ID135, located in the non-fractured east compartment show only some fractures with isolated microfissures : the filling is thin with calcite.

Investigations based on different diagrams in the boreholes have perfectly highlighted the presence of these two quite contrasting compartments. Regarding porosities for example, it is possible to delimit zones which could be favourable for any fluid transfer from the aalenian aquifer.

After several months, following the drilling operations, the presence of fluid was detected in the four boreholes ID0, ID225, ID270 and ID315 which are the most strongly affected by the disturbed zone. Flow rates from 5 to 10 ml/h have been detected from boreholes ID0 and ID315. Video inspections into these wells have enable the fractured levels, from which the fluids seem to emanate, to be precisely located : these are the zones displaying the most evidence of tectonics. The proximity of the aalenian aquifer (one tenth of a meters from the well end ID0), associated with the drilling of the borehole itself, could explain this delayed fluid production from the formation in this particular context.

**General Organisation of the Fracturation**

Thus, at this detailed scale, one can verify that the whole rock formation is affected by a fracturation network characterised by three main families of planes [1, 2] : a strongly inclined NNW-SSE family, which is the most representative, a nearly E-W family and a NE-SW family. These families have been identified at the surface, and are better known thanks to the detailed studies from the cores of the drillings into the tunnel.

As has been observed on the different sites at the surface at the same stratigraphic levels, fracturation exhibits two main kinds of plane : planes with calcite fillings (of microscopic to centimetric thickness), and planes without any filling. On the level of the major planes (category C1), the nearly N-S direction predominates, as is the case at the surface. With the second category (C2), fracturation
seems to be organised according to three well-defined plane families: close to N-S, close to E-W and NE-SW. However, in the small planes category (C3), fracturation presents a wide azimuthal variation. These observations emphasise the necessity to analyse fracturation by considering its relative scale, because all the fractures do not have the same importance: this is fundamental with regard to the relative plane influences on the permeability characteristics of a formation.

Fracturation Influence on In Situ Permeability Tests

Six permeability tests were performed [3] within the argillaceous toarcian formation from the ID180 borehole, each test including a long stabilisation phase (more than 24h), and a transient phase after a hydraulic pulse. One test, performed in a single 100 m long test room from the well bottom indicates a very low permeability value of approximately \( 1.4 \times 10^{-7} \) m/s. Permeabilities values, obtained from tests with double packers room (1.50 m) situated in zones having well documented fractures appear slightly higher: between \( 1.3 \times 10^{-7} \) m/s and \( 6.7 \times 10^{-7} \) m/s. The interpretation of the data from these comparative hydraulic in situ tests indicates that in the worst cases, the increase in permeability values that might be ascribed to the presence of fractures would be of about one order of magnitude: \( 1 \times 10^{-9} \) to \( 1.1 \times 10^{-7} \) m/s for the fractured zones, and \( 1 \times 10^{-10} \) to \( 1.1 \times 10^{-10} \) m/s for the clayey matrix itself. This does not reveal any really significant differences.

TEXTURAL, MINERALOGICAL AND GEOCHEMICAL STUDIES OF THE FILLINGS

Aim of the Study

In order to characterise better the potential transfers of water through the fractures of the Toarcian and Domerian claystones, a detailed study of the filling material of the fractures has been scheduled. Textural, mineralogical, geochemical and isotopic informations related to the fracture fillings are indeed essential to identify paleocirculations, to classify the successive generations of fractures (during compaction, cementation and tectonic events), and to distinguish among them those which might have acted as privileged pathways for water circulations on long distances. In addition, the knowledge of the chemical nature of the paleofluids, particularly the role of the matrix for the regulation of fluid chemistry through water-rock interactions, and some insight in the mechanisms of fracture sealing represent also an important aspect of the study as long as element migration and safety assessment are concerned.

Several and complementary microscopy and spectroscopy analysis have been performed in this context. The scope of this paragraph is not to draw definitive conclusions, but rather to communicate some preliminary results. The study about the natural isotopes of the fracture fillings is presented in the next paragraph. The samples come from boreholes compartment (ID0, ID180, ID225, ID270 and ID315) corresponding to the western, that exhibits numerous fissure or fracture type structures. Twenty thin, polished and oriented sections have been made. The nature of the mineral paragenesis and the chemical contents of the fracture fillings have been determined through standard thin-section petrography, cathodoluminescence microscopy, scanning electron microscopy, electron microprobe and ICP-MS. In addition, fluid inclusions present in the calcite of the fracture fillings have also been considered. To appreciate the matrix influence on the chemistry of the mineralising fluids, the mineralogy and the chemical contents of the rock matrix surrounding the fractures have been made via ICP-AES, ICP-MS and standard mineralogical techniques. In complement, a cartography of the uranium distribution on thin sections containing both matrix and fractures have also been performed to evaluate the uranium leaching and migration through fractures.
First Results

The fracture fillings are mainly constituted of calcite. Four types of filling paragenesis have been identified: 1) calcite alone, 2) calcite and framboidal pyrite, 3) calcite and cubic pyrite, and 4) calcite and barite. The morphology of calcite crystals is very variable, even within a same fracture. Elongated and rhomboedric crystals are the most common observed morphologies. The interface between matrix and fissures or fractures is sometimes clean, but most of the time it is diffuse with a dense stockwork of microfissures or with matrix pieces trapped in calcite.

Some of the results concerning the fracture and matrix chemistry are plotted following a stratigraphic profile in figure 6. A negative correlation seems to occur between the carbonate content and the concentration of major and trace elements in calcite fillings. The « stratigraphy » influence is well correlated to the transition from marls to argilites. The hypothesis of regulation of the matrix seems to be reinforced by the preliminary results coming from REE and 87Sr analysis. The matrix influence could be related to the very low solution/rock ratios due to the matrix microporosity, to the size of the fractures, and to the presence of dense stockworks of microfissures privileging fluid circulation at local scale. Local migrations of interstitial fluids and pCO₂ drops in open fracture could explain most of the fracture sealing. Several indices point out very slow water circulations: 1) the scarcity of the fluid inclusions, 2) the presence of pyrite and calcite macrocrystals, and 3) a matrix signature of the fracture fillings. The scarcity of fluid inclusions has not allowed to specify a range of temperatures for the calcite crystallisations in the fractures. The cartography of the uranium distribution show no specific uranium leaching from matrix and no uranium migration through fractures. These results, in addition with high iron contents in the calcite fillings, suggest the circulation of reducing fluids within the fractures.

The numerous textural, mineralogical and geochemical elements have now to be related to the fracturation organisation data to attempt a classification of the fractures regarding the diagenesis and the paleocirculations. The existence of some anomalies in chemical contents, by comparison to the general stratigraphy trends, still remains to be interpreted.

ISOTOPIC DATA FROM CARBONATES FROM FRAC TURES

Fractured zones which were analysed are mainly located in the western part of the study sector [7]: boreholes ID0, ID180, ID225, ID270 and ID315. For boreholes CD et CA, fracturation is situated only in some sectors [8] that can be lumped into seven main zones numbered n°1 to n°7.

\(^{13}C\) And \(^{18}O\) Contents : Origin of the Carbonates from Fractures

When precipitation takes place at equilibrium, \(^{13}C\) and \(^{18}O\) contents of the solid carbonates are a function of temperature. \(^{18}O\) content of the water and of the \(^{13}C\) content from the Total Inorganic Dissolved Carbon (TIDC). They can be used to retrace some of the fluid characteristics from which they derive. The data discussed are presented figure 7: diagram \(^{13}C\) vs \(^{18}O\), and respective profiles of the \(^{13}C\) et \(^{18}O\) of these minerals vs depth figure 8. Data concerning \(^{13}C\) and \(^{18}O\) isotopic contents from the matrix are reported too as reference and for comparison.

In the \(^{13}C\) vs \(^{18}O\) diagram the increase in the \(^{13}C\) content from secondary calcites from the upper part of the series (\(^{13}C\) = -3 to -6 \(^{\delta}_{\text{PDB}}\) for \(^{18}O\) content not different from -6\(^{\delta}_{\text{PDB}}\)) corresponds to the fillings close to the Aalenian aquifer (borehole CA) : this could indicate a simple reservoir effect, where the fluid which percolates is progressively enriched in \(^{13}C\). Precipitation of a biogenic CO₂ in the TIDC of the solution could logically explain these very negative content values at the top of the argillaceous series.
Representative data from carbonates of fractures localised into the upper Toarcian and Domerian fit quite well with two parallel lines (slope 0.7). The $^{13}$C difference of 1.3 $\%$ between the two lines could be the result either of a temperature difference between two types of fluids (approximately a 10$^\circ$C difference), or in a variation in the $^{13}$C content from the total inorganic dissolved carbon of the initial solution. But this splitting into two families cannot be linked neither to the situation of the samples in the series, nor to what is known from the different fracturation phases at the present time.

Minerals found deeper in the series (borehole ID180) are richer with $^{13}$C (positive values from 0 to +2 $\%$ vs PDB). They could be in equilibrium with the carbonated phase of the matrix (approximately 0.5 $\%$ vs PDB). Samples with $\delta^{18}$O values between -8 to -10 $\%$ vs PDB for the high $\delta^{13}$C values ($\approx$ 1 $\%$ vs PDB), are representative of samples localised not far from the lower Toarcian ("Schistes Carton", boreholes CD).

The two profiles of $^{13}$C et $^{18}$O contents from these minerals vs depth (cf. figure 8) confirm the above statements.

The monotonous profile of $^{18}$O with depth shows a progressive depletion from the top to the bottom of the series which could result from simple thermal effects and/or from fluctuations of the isotopic composition of the initial fluid.

Differences in the temperatures of the deposits calculated on the basis of the interstitial pore water $^{18}$O content will imply a geothermal gradient higher than the normal local situation, considering that all the secondary calcites have precipitated from the same original solution (which is not proved).

$^{18}$O contents changes within a same fractured zone could be compatible with re-crystallisation processes, with a fluid with isotopic contents different from those of the initial fluid [5, 9].

We may well envisage percolation of fluids of different isotopic contents and different temperatures over time. Thus we may admit the existence of two « groups » of carbonates from fractures, with different $^{18}$O mean contents values [8]:

- a first group (fractured zones 1, 3 and 4), corresponding to $^{18}$O contents higher than the one of the aalenian aquifer, or corresponding to temperatures close to the present annual mean temperature;
- a second group (fractured zones 5, 6 and 7), corresponding to $^{18}$O contents similar to the one of the aalenian aquifer, or corresponding to temperatures higher than the present annual mean temperature.

Although $^{13}$C contents from the calcites can differ by about 1 $\%$ vs PDB within a same zone of fracture, these $^{13}$C evolve between the Aalenian to the upper Domerian describing a kind of bow, with a regular tendency towards enrichment in the heart of the formation. It is impossible to explain simply such a profile by temperature alone. This particular evolution of $^{13}$C contents may be explained through to several hypothesis, presupposing both upwards and downwards circulations.

This general distribution of the $^{13}$C contents could reflect a progressive increase of exchanges with the matrix, and/or of the pH, from the extremities of the profiles toward the centre of the Toarcian formation. This hypothesis [8] of a mixture between a solution of meteoric origin (containing partly biogenic carbon) and a connate solution (containing sea carbonates) may be considered in so far as the fracture presenting highest $^{13}$C contents are located in the profile in immediate vicinity of zones where rather high $^{18}$O and $^3$H contents from interstitial waters have been found.

Considering a possible correlation between structural plane characteristics and isotopic contents it appears that the nearly E-W planes (C2 category) present the highest $\delta^{18}$O content (-6.5 to -6 $\%$ vs PDB) and the nearly N-S sub-vertical planes (main C1 category) correspond to values slightly lower ($\delta^{18}$O = -8 to -6.5 $\%$ vs PDB).

$^{87}$Sr/$^{86}$Sr isotopical studies could give complementary informations on the origin of the carbonates fillings. For the moment, a too small number of data has been collected to draw any conclusions.
Nevertheless, with the exception of two small variations, \(^{87}\text{Sr}^{86}\text{Sr}\) distribution values seem to be very uniform (ranging from 0.70841 to 0.70858) for the fractures present in argilites.

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**C and U/Th Contents: Dating of the Carbonates from Fractures

**C Contents

The dating method using radiocarbon theoretically covers a time period from 0 to 60,000 years, this latter being the upper limit due to the period of \(^{14}\text{C}\) (5730 a). Five carbonated fillings from fractures of boreholes CA and CD were subjected to a \(^{14}\text{C}\) test using either \(\beta\) spectrometry or mass spectrometry by particle accelerator.

All the tested samples present activities lower than 0.31%, meaning "apparent ages" higher than 40,000 years. If we consider that 50% of the carbon comes from the matrix (activity equal to 0%) and that 50% comes from meteoric water (activity equal to 100%), it can be estimated that no important circulation has occurred during at least the past 20,000 years.

**U And Th Contents

U/Th contents measurements using mass spectrometry by thermo-ionisation (T.I.M.S., GEOTOP - University of Quebec, Montréal) were performed on five samples from fillings of fractures from boreholes CA and CD. Variations of the Th/U ratio from a sedimentary medium of relatively homogenous mineralogical composition can be the sign of uranium mobility, implying the existence of percolation of oxidising solutions. Such mobilisations result in a disequilibrium in the \(^{230}\text{Th}^{234}\text{U}\) and \(^{234}\text{U}^{238}\text{U}\) activities ratios for which the time of return to equilibrium continues to be measurable several hundred thousand years for the first, and around two millions years for the second.

Uranium and thorium contents are quite very variable from one sample to another, with a strong tendency to decrease with depth, and the presence of a very large detrital fraction (matrix) identified because of the \(^{232}\text{Th}\) concentration. Samples with this high \(^{232}\text{Th}\) contents levels correspond to the highest \(^{238}\text{U}\) contents in the serie: uranium present in these fractures were therefore essentially brought by the detrital fraction. Consequently, the very low quantities of uranium incorporated via chemical processes into the carbonates of fractures during precipitation suggest that the initial solutions were poor in dissolved uranium, indicating a rather low redox potential.

Two samples located into the upper part of the clayey series present low but significant disequilibrium of the two isotopic ratios \(^{234}\text{U}^{238}\text{U}\) and \(^{230}\text{Th}^{234}\text{U}\). These disequilibria could indicate uranium mobilisation phases during these last two million years. Furthermore, a very high \(^{230}\text{Th}^{234}\text{U}\) ratio for one sample located into the lower part of the series (borehole CD) could indicate that a loss of uranium may well have taken place during remobilisation phase due to circulation later than to this deposit and more recent than 600,000 years but considering the very small \(^{230}\text{Th}\) content, this result has to be confirmed.

**SUMMARY AND CONCLUSIONS

The fracturation analysis of the Domerian and Toarcian argilaceous serie at Tournemire performed from fourteen boreholes has revealed the existence of different plane families with a main NNE-SSW one which constitutes the principal paleocirculations network within the Tournemire massif.

Fracturation from cores features two kinds of planes: planes with different types of calcite fillings (microscopic to centimetric), and planes without fillings. This fracturation seems perfectly sealed and impervious to any circulation. Nevertheless, drillings close to the upper aquifer in a disturbed zone seems to have brought about fluid production several months after the work had ended.

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In situ permeability pulse-tests in the boreholes targeting previously identified fractured zones have yielded local permeability values slightly higher than the « global matrix permeability » measured in one long room (from $< 10^{-13}$ to $< 10^{-14}$ m/s).

Texture, mineralogy, chemical and isotopic contents of the secondary carbonates from the fractured zones have been analysed in order to determine the origins, ages and chemistry of the initial circulations. Four types of filling paragenesis have been identified: calcite alone, calcite and framboidal pyrite, calcite and cubic pyrite, and calcite and barite. The morphology of calcite crystals is very variable, even within a same fracture. The « stratigraphy » influence on fracture filling chemistry through water-matrix interactions seems strong. Several indices point out very slow water circulations: the scarcity of the fluid inclusions, the presence of pyrite and calcite macrocrystals, and a matrix signature of the fracture fillings. The scarcity of fluid inclusions has not allowed to specify a range of temperatures for the calcite crystallisations in the fractures. The cartography of the uranium distribution show no specific uranium leaching from matrix and no uranium migration through fractures. These results, in addition with high iron contents in the calcite fillings, suggest the circulation of reducing fluids within the fractures.

The $^{18}$O content profiles of secondary carbonates cannot be justified by thermal effects only, because the calculated temperature gradients are abnormally high considering regional tectonics situation. Thermal effects cannot fully explain too the spacial variability of the $^{18}$O content distributions: both different isotopic contents and temperatures evolving over time may be envisaged for the percolation fluids. This hypothesis would tend to indicate the coexistence at least of two « groups » of fillings corresponding to different isotopic contents and temperatures.

Interpretation of $^{13}$C content with depth is more complicated, and could necessitate having recourse to hypotheses involving leaching of the matrix specially in the centre of the argillaceous formation. These hypotheses imply fluid circulations both upwards and downwards. Unfortunately this have not been resolved by using $^{14}$C dating and U and Th family disequilibria: $^{14}$C activity close to $\pm 0 \%$, and $^{234}$U/$^{238}$U often close to unity.

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Figure 1: Map of the structural elements in the vicinity of Tournemire (Aveyron - France)
Figure 2: Geological cross-section and block-diagram with boreholes position at the Tournemire tunnel
### Figure 3: Fracture localisation, developed cores. Wulff stereographic representations.

Categories of planes C1, C2, C3 and stratification planes.
Figure 4: Fracturation in the ID boreholes sector

Figure 5: Fracturation characteristics from boreholes
Figure 6: Matrix and fracture chemistry following a stratigraphic profile.
Figure 7: δ¹⁸O vs δ¹³C (°/vs PDB) - Carbonates from fractures and matrix

Figure 8: δ¹⁸O and δ¹³C (°/vs PDB) vs depth (equivalent stratigraphy) - Carbonates from fractures and matrix
A strontium and lead isotopic study of the Marcoule silty-clay layer (Gard, France) and implications for characterisation of water-rock interactions in low permeability argillaceous media.

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Abstract

The evolution of Rb/Sr and U/Pb isotopic systems is commonly used by geochemists for dating and tracing geological processes. In this study, Sr and Pb radiogenic natural tracers have been applied to sedimentary rocks recovered from five ANDRA boreholes. These boreholes were drilled into Cretaceous layers of the South-East sedimentary basin and led to the discovery of the "Marcoule silty-clay layer" (MSL), a Vraconian silto-argillaceous formation, composed of homogeneous and low permeable sediments, rich in detrital muscovites and authigenic pyrites. This formation is bounded by two aquifers. The main aquifer is located in the overlying Cenomanian sediments; the other one is restricted to a thin sandstone level located between the MSL and Urgonian limestones. Since Cretaceous times, the studied area might have undergone tectonic and morphological changes through the onset of the Pyrenean and Alpine orogenies, the Oligocene distension, and the Messinian phase. Such events were likely to produce structural discontinuities, fluid flows and water-rock interactions into the MSL.

The aim of this study was to determine if Rb/Sr and U/Pb isotopic tracers remained closed geochemical systems in the MSL, during the last 100 Ma. As these elements are particularly mobile when exposed to fluid flows in sedimentary basins, any loss or gain in studied radioactive and radiogenic isotopes will induce variations on the measured isotopic compositions and will reveal water-rock interactions. In addition, Rb/Sr and U/Pb tracers provide good natural analogues of some radionuclides present in radioactive wastes (Cs (Rb), Ra (Sr) and intermediate daughters of U).

110 Rb/Sr and U/Pb analyses of whole rock and mineral samples were conducted. In Rb/Sr and U/Pb isochron diagrams, experimental points of MSL rocks define linear arrays which result from a binary mixing between authigenic or diagenetic components (carbonate, pyrite) with low radiogenic Sr and Pb isotope compositions and a detrital highly radiogenic end-member composed of phylrites and in particular of muscovites derived from the Variscan micaschists of the Cévennes. A slight and gradual increase of $^{87}\text{Sr}/^{86}\text{Sr}$, $^{87}\text{Rb}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios occurs from carbonate-rich levels at the bottom of the MSL to more detrital ones upwards. However discontinuous variations in Sr and Pb isotope ratios are never recorded in the unfractured MSL rock samples of the 2 boreholes (MAR 202 and 203) located near the Marcoule Nuclear Center.

In addition, 18 whole rock and mineral (calcite, celestite, pyrite) samples were analysed in the vicinity of a fault crosscutting the MSL in order to define the Sr and Pb isotopic anomalies, generated in the MSL by water-rock interactions. This fault, found at -600 m in depth (MAR 501 borehole) is one of the submeridian faults affecting Cretaceous sedimentary formations in the Bagnols-sur-Cèze area (Gard, France).
In the studied fault zone, calcite and celestite exhibit $^{87}$Sr/$^{86}$Sr ratios equilibrated with the Sr composition of diagenetic carbonates. However, $^{206}$Pb/$^{204}$Pb ratios of authigenic pyrites, disseminated within MSL, are distinctly less radiogenic than those of pyrites present with calcite and celestite crystals in tension gashes and in the tectonic breccia of the fault zone. These Pb data suggest a fluid paleocirculation related to the beginning of the Oligocene distensive phase. In the MSL whole rocks, such a paleocirculation produces a discontinuous Sr isotope anomaly related to secondary calcite and celestite enrichment in the vicinity of the fault. But at a smaller scale, secondary calcite and celestite-free MSL whole rock samples collected along the fault zone, never record any Sr isotopic evidence of water-rock interaction, which is in good agreement with the very low permeability of MSL sediments.

Drilled by MAR 202 and 203 boreholes, unfractured MSL whole rocks do not exhibit discontinuous Sr and Pb isotopic anomalies and yield strong evidences for an evolution of Rb-Sr and U-Pb tracers as closed systems i.e. without interaction with a fluid, in the past or at the present time.

INTRODUCTION

In geochemistry, Rb/Sr and U/Pb isotope variations are commonly used for dating and tracing geological processes and formations [1]. Basically, the decay with time of a radioactive father isotope P, as $^{87}$Rb, $^{238}$U or $^{235}$U, produces a radiogenic daughter isotope F, as $^{87}$Sr, $^{206}$Pb or $^{207}$Pb. Variable abundances of F in rocks and minerals are normalized to the constant abundance of an unradiogenic isotope F', as $^{86}$Sr or $^{204}$Pb, of the same element. A genetic suite of rocks of magmatic or high grade metamorphic origin, crystallized at high temperature, shows variable Rb/Sr and U/Pb ratios and an initial isotopic homogenization of Sr and Pb. Thus such rock samples will plot as points on a straight line parallel to the x-axis, in the so called "isochron diagram" (Fig. 1a). After radioactive decay with time, experimental points define a linear array called isochron. This crystallization age of the analysed samples is given by the slope of this isochron and their initial ratio by its y-intercept. Meaningful crystallization ages are obtained insofar as the studied samples behaved through time as closed systems with respect to P and F. In that case, P and F abundances are exclusively a function of radioactive decay. Thus, isochron diagrams are usually difficult to be used for dating sedimentary rocks formed by the mixing of various detrital, authigenic and diagenetic components which are not isotopically homogeneized. In addition, Rb, Sr, Pb and especially U are mobile during water-rock interactions in sedimentary basins. A suite of rocks may be also formed by mixing of two components with different P/F and F/F' ratios, which will provide a fictitious isochron (in fact a mixing line). Such cases are known for magmatic rocks ; the assumption has been done [2,3] of mixing line occurrence in isochron diagrams for suites of sedimentary rocks, when their Rb/Sr (or U/Pb) systems are controlled by two main components with very different P/F and F/F' ratios (Fig. 1b).

GEOLOGICAL BACKGROUND

In France, the South-East sedimentary basin is bounded to the west by the southern Massif Central and to the east by the Alpine mountain belt. More than 8000 m thick, this basin constituted the continental shelf of the Thetis ocean during Jurassic and Cretaceous times. Between the inactive Cévennes fracture zone and the present-day active Nîmes fault (Fig. 2a), the 100 Ma old Marouelle silty-clay layer (MSL) have been discovered from -400 to -800 m in depth, by the ANDRA MAR 202 and 203 boreholes (Fig. 2b). Overlying Urgonian limestones, this Upper Albian formation (Vraconian in terms of local geology) is made of very homogeneous, unfractured and low permeable silty clays (2.6 $\times$ 10$^{-14} < K < 5.7 \times 10^{-12}$ m/s). Composed of quartz (30-50 %), smectite clays (30-40 %) and carbonates (10-20 %) this silt, contains in addition detrital muscovites and authigenic pyrites. Some glauconite-rich levels are encountered in the upper part of the MSL.

Although remarkably homogeneous, this formation shows a slight and gradual geochemical evolution from carbonate-rich levels at the bottom to more detrital ones upwards ; to
the NW, when approximating the margin of the basin, its becomes thinner and is crosscut by Cretaceous submeridian faults, recognized at -555 m and -600 m in depth by the MAR 501 borehole (Fig. 2). Further North, the MSL deepens and its thickness decreases down to 150 m, according to stratigraphic logs yielded by MAR 401 and 402 boreholes (Fig. 2a). The MSL is bounded by two aquifers. The main upper aquifer is located into Cenomanian sandstones, interbedded with clays, limestones and lignites (Fig. 2b). A minor lower aquifer is restricted to a thin sandstone layer overlying the Urgonian limestones (Fig. 2b). Since Mesozoic time, the studied area might have undergone tectonic and morphologic changes through the onset of the Pyrenean orogeny, the Oligocene rifting, the Alpine orogeny and the Messinian phase. Such geological events were likely to produce faults, fluid flows and water-rock interactions, in the Marcoule silty-clay layer.

RESULTS AND DISCUSSION

MSL mixing lines in Rb/Sr and U/Pb isochron diagrams

The systematics of Rb/Sr and U/Pb isotopic evolution for rock and mineral samples of MAR 202, 203, 402 and 501 boreholes were attempted by Lancelot et al. [6,7,8,9]. Their results for MSL whole rocks are shown in the Rb/Sr and U/Pb isochron diagrams (Fig. 3); 31 MSL drilled samples and 3 Gargasian marls collected on the field were analysed by Rb/Sr method and define a linear array A whose slope indicates a date of 251 ± 17 Ma and an initial 87Sr/86Sr ratio of 0.70749 ± 0.00008. Taking into account the Rb and Sr contents and the present-day 87Sr/86Sr ratio of each sample, initial 87Sr/86Sr and 87Rb/86Sr ratios were calculated back to 100 Ma; plotted in the Rb/Sr isochron diagram (Fig. 3), they yield an initial linear array A₀ whose slope indicates a date of 154 ± 14 Ma and an initial Sr composition of 0.70747 ± 0.00007.

The residence time of Sr in the oceans is about 5 Ma which is much longer than the seawater mixing time of some thousand years and variations of seawater 87Sr/86Sr ratio over geological time is well documented [1,2,8,9,10,11,12,13,14,15]. The high-resolution seawater Sr isotope curve [16] generated through the analysis of well-dated and well preserved fossils indicates a variation of seawater 87Sr/86Sr ranging from 0.707315 to 0.707415 between 110 Ma and 97 Ma (Gargasian-Vraconian). In the studied area, additional Sr determinations were performed by acid leachings [6], both on calcite of Gargasian belemnite guards and on Gargasian and Vraconian carbonates. The first ones suggest a 87Sr/86Sr value of 0.70732 ± 0.00003 for the South East basin seawater during Gargasian times in good agreement with Jones et al.'s conclusions [16]. The second ones provide a mean value of 0.70747 ± 0.00006 (0.70741 ± 87Sr/86Sr ≤ 0.707578) for diagenetic carbonates, crystallized in Gargasian marls and Vraconian silts which led to conclude that the initial Sr ratio of 0.70747 ± 0.00007 provided by the y-intercept of the linear array A₀ in the Rb/Sr isochron diagram (Fig. 3) represents the isotopic composition of diagenetic carbonates.

In the Rb/Sr diagram, when experimental points define a linear array with a positive slope it corresponds either to an isochron or to a mixing line; then, all the analysed samples are likely to be synchronous and the value of the age of the sediment deposition (= 100 Ma), whereas the y-intercept of the linear array provides the Sr isotope value of diagenetic carbonates of the MSL. The linear array may therefore be interpreted as a mixing line. As quartz and pyrite present in MSL rocks are very depleted in Rb and Sr, mixing processes are believed to involve two main components: a carbonate phase as the low radiogenic Sr end-member and Rb-bearing muscovites and clays as the high radiogenic one.

The date of 251 ± 17 Ma indicated by the slope of the mixing line represents the addition of the age of deposition of the sediments (i.e. 100 Ma) with the apparent age yielded by the initial mixing line (154 ± 14 Ma). Gargasian marl and Marcoule silty clay points plot on the same mixing line which led to conclude that there is no variation, during Gargasian and Vraconian times, of the radiogenic end-member i.e. the detrital muscovites. The homogeneity of this muscovite component suggests a detrital flux from a muscovite-rich homogeneous metamorphic basement outcropping during mid-Cretaceous times on the margins of the basin. From this point
of view, the fairly uniform Variscan micaschist series of the Cévennes (Fig. 2) dated at 335 ± 5 Ma [17] might be a good candidate for the source of the studied sediments. $^{39}$Ar/$^{40}$Ar dating of MSL detrital muscovites, point by point, is planned to confirm this assumption.

Plotted in the U/Pb isochron diagrams, 14 MSL drilled samples, 3 Gargasian marls, 1 carbonate fraction and 2 framboylite pyrite fractions define a linear array whose slope indicates a date of 150 ± 13 Ma and an initial $^{206}$Pb/$^{204}$Pb ratio of 18.65 ± 0.005 (Fig. 3). Initial U/Pb linear array has been also calculated back to 100 Ma; its slope indicates a date of 57 ± 10 Ma and its y-intercept ($^{206}$Pb/$^{204}$Pb = 18.645 ± 0.005) is interpreted as the Pb composition of the mid-Cretaceous seawater from the South East basin, recorded in authigenic framboylite disseminated pyrites.

$^{87}$Sr/$^{86}$Sr evolution with time for Marcoule mid-Cretaceous sedimentary formations

Using the same analytical data that for the isochron diagram, it is useful in describing the increase of $^{87}$Sr/$^{86}$Sr ratios as a function of time in a suite of cogenetic rocks. For each rock, the Sr evolution is represented by a straigh line whose slope is ($^{87}$Rb/$^{86}$Sr)$\lambda$. Figure 4 is such a Sr evolution diagram for MSL, Cenomanian and Aptian whole rocks from MAR 202, 203 and 501 boreholes. Unlike a suite of comagmatic rocks which commonly have the same initial Sr ratio, studied sedimentary rocks have different ($^{87}$Sr/$^{86}$Sr)$_0$ ratios (Fig. 3) caused for example by variation in the amount of unradioactive carbonates and radiogenic muscovite (and clays) in MSL samples. The Sr evolution diagram shows that the MSL domain is obviously smaller than those of the overlying and underlying formations. The Sr heterogeneity of Cenomanian and Aptian rocks is related to their variable petrology and also to water-rock interactions in the two aquifers. At the opposite, Marcoule silty-clays show very homogeneous evolution of Sr with the time, which means rather similar $^{87}$Sr/$^{86}$Sr and $^{87}$Rb/$^{86}$Sr ratios. Nevertheless, in MAR 202 and 230 boreholes, a slight and gradual increase of these ratios is observed from the bottom to the top of the MSL. This pattern is due to detrital Rb-bearing minerals slightly more abundant in MSL upper layers than in carbonate-rich lower ones.

The isotopic homogeneity of the Marcoule silty-clay layer

The contrast between the isotopic homogeneity of MSL and the heterogeneity of overlying and underlying formations, is also clearly demonstrated by depth profiles of $^{87}$Sr/$^{86}$Sr, $^{87}$Rb/$^{86}$Sr, $^{206}$Pb/$^{204}$Pb and $^{238}$U/$^{204}$Pb ratios in MAR 202 and 203 boreholes (Fig. 5). These isotopic ratios concern elements (Rb, Sr, U) very mobile in the upper continental crust during water-rock interactions. In addition, $^{87}$Sr/$^{86}$Sr and $^{206}$Pb/$^{204}$Pb ratios directly reflect gain or loss of radioactive $^{87}$Rb and $^{238}$U which are among the most leachable isotopes by oxidizing waters [18,19,20]. For the 400 metres thick MSL, the four isotope ratios are very similar and do not show local discontinuous variation (Fig. 5), which suggest both a rather homogeneous chemical composition and Rb/Sr and U/Pb isotopic evolutions as closed systems. At the opposite the overlying Cenomanian or underlying Aptian formations show great discontinuous variation of the four ratios (Fig. 5) related both to petrological heterogeneities (for example Cenomanian interbeddings of sandstones, limestones, clays and argillaceous limestones (Fig. 2b)) and to water-rock interactions in the upper and lower aquifers. In addition, present-day $^{87}$Sr/$^{86}$Sr and $^{87}$Rb/$^{86}$Sr ratios of MSL rocks are more radiogenic in MAR 501 borehole than in MAR 202 and 203 ones (Fig. 5), in agreement with the increase of detrital Rb-bearing minerals in the MSL towards the margins of the South-East basin.

Pb/Pb dating of a paleofluid flow

Authigenic spheroids of octahedral pyrite crystals are common in the unfractured Marcoule silty-clays. These spheroids are individualized or coalescent, coating detrital grains; for comparison, identical pyrite spheroids were extracted from oozes younger than 100 years ($^{210}$Pb dating [21]) of the Thau lagoon (Hérault, France). In the MAR 501 borehole, at -600 m in depth, a fault crosscuts the MSL (Fig. 2b). In this fault or in its vicinity calcite, celestite, barite and cubic
pyrite crystallized in the cement of the fault breccia and in vertical tension gashes, 0.1 to 2 millimetres width.

Plotted in the $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram (Fig. 8), Pb isotope compositions of MSL pyrite spheroids are less radiogenic than MSL cubic pyrites. Gargasan carbonates and disseminated pyrite spheroids from Gargasan marls and Vracanian silty clays, show similar Pb isotopic ratios (Fig. 8). The local Pb isotopic composition of the Cretaceous seawater in the South East basin, is well recorded in these authigenic or early diagenetic components. The radiogenic Pb of cubic pyrites, crystallized in tension gashes and fault breccia, would represent the mean isotopic composition of the lead extracted from the brecciated MSL rocks by water-rock interaction at the time of the fluid flow along the fault. According to the Stacey-Kramers model [22], the Pb isotopic evolution defined by the two generations of pyrites in the MSL, is best described by a secondary lead growth curve with a $\lambda_2$ value of 9.81. The isotopic discrepancy observed between spheroidal and cubic pyrites then corresponds to a time interval of 58 Ma. Assuming an authigenic or early diagenetic origin for the MSL disseminated pyrite spheroids ($t = 100 \pm 10$ Ma), a crystallization age of about 42 $\pm$ 10 Ma may be calculated for the cubic pyrites. Recent Pb/Pb data on granitic rocks, K feldspars and Pb-Zn ores of southern Sardinia (Italia) and southern french Massif Central [17,23,24,25] allowed to define a regional Pb/Pb growth curve for these two crustal segments of the Variscan belt (Fig. 8). According to this two-stage model and considering that the MSL is derived from the Variscan basement of the southern Massif Central, Pb/Pb ages of 86 Ma and 39 Ma are obtained for each generation of the MSL pyrites in agreement with the previous age calculation (S.-K. model). In conclusion, along the studied fault of the MAR 501 borehole, a water-rock interaction occurred in the Marcoule silty-clay layer 40 Ma ago ; thus, calcite and pyrite have crystallized in tension gashes and fault breccia at the end of the Pyrenean compressional event or at the beginning of the Oligocene rifting episod.

Sr isotopic anomaly generated by a fluid flow

Isotopic variations induced by fluid flows through the MSL, were studied in detail by additional Sr data performed on whole rocks and mineral fractions (celestite, calcite). Whole rock samples were collected in the vicinity of the fault studied for Pb isotopes:
- at about 5, 10, 15, 20 and 25 m of the fault limbs,
- up to 20 cm of the fault, but with or without tension gashes filled with calcite, barite, celestite and pyrite,
- in the fault zone (one MSL clast and one slickenside sample).

Sr analyses were also performed on celestite crystallized as needles in the cement of the fault breccia, and on calcite extracted from this cement and from tension gashes up to 20 cm of the fault limbs.

MSL whole rocks sampled from 5 m to 25 m of the fault, show $^{87}\text{Sr}/^{86}\text{Sr}$ ratios ranging from 0.70902 to 0.70973. No increase (or decrease) in these Sr ratios, is observed approximating the fault zone (Fig. 6a and 7). Plotted in the isochron diagram (Fig. 3), these whole rocks define with the other MSL samples of MAR 202 and 203 boreholes, the present-day mixing line previously described. A MSL whole rock (WR1), sampled at few centimetres of the upper limit of the fault, provides a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70929 in the range of the Sr values found for other MSL rocks sampled from 5 m to 25 m of the fault. This sample does not exhibit tension gashes filled with calcite, celestite and pyrite. At the opposite, MSL whole rocks with such tension gashes, sampled above (WR2) or under (WR4) the fault, yield less radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.70824 and 0.70844 respectively (Fig. 6a). Celestite and calcite fractions extracted from the cement of the fault breccia and from tension gashes, show very low Sr isotopic compositions ranging from 0.70749 to 0.70768. Thus, in the vicinity of the fault, the occurrence of tension gashes, filled with calcite, celestite and barite produces a discontinuous variation of Sr isotopic compositions in the MSL whole rocks (Fig. 6a and 7). Comparison of Sr isotope ratios of these calcites and celestites with the Cretaceous diagenetic carbonates (Fig. 7) suggests that the fluid responsible for the calcite and celestite crystallization had mainly isotopically equilibrated its strontium with such Cretaceous carbonates.

The Sr study of the fault was achieved by the analyses of a slickenside sample (WR5) and of a MSL clast (WR3) from the fault breccia ; they respectively provide similar $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.70886 and 0.70893. These samples are less radiogenic than MSL whole rocks (without calcitic gashes) sampled at the same depth (Fig. 7) which suggests that a small interaction with the fluid
responsible of the calcite crystallization in open tension gashes, is recorded in WR5 and WR3 samples.

CONCLUSIONS

1/ MSL whole rock samples from MAR 202, 203 and 501 boreholes, define mixing lines, both in the Rb/Sr and in the U/Pb isochron diagrams. A binary mixing is assumed between low radiogenic components (diagenetic carbonates for Sr, authigenic or early diagenetic pyrites for Pb) and high radiogenic ones (Rb-bearing clays and detrital muscovites inherited from a homogeneous source, i.e. the 335 Ma old micaschists of the Cévennes Variscan basement). In the Rb/Sr isochron diagram, the Y-intercepts of the mixing line, provide the Sr isotopic composition of diagenetic carbonates.

2/ A fault crosscutting a MSL core of the MAR 501 borehole has been intensively studied. Located in the fault breccia or filling tension gashes surrounding the fault, calcite, celestite and barite crystallized in relationship with the paleocirculation of a fluid along the fault within indurated MSL sediments. Celestite and calcite samples yield homogeneous $^{87}$Sr/$^{86}$Sr ratios ranging from 0.70749 to 0.70768, mainly isotopically equilibrated with Cretaceous diagenetic carbonates. In the MAR 501 boreholes, studied MSL whole rocks usually show $^{87}$Sr/$^{86}$Sr ratios ranging from 0.70902 to 0.70973 but the presence of the fault is outlined by a local high Sr anomaly (0.70824 $\leq$ $^{87}$Sr/$^{86}$Sr $\leq$ 0.70844) related to calcite and celestite occurrence in tension gashes. At a smaller scale, a calcite and celestite-free MSL whole rock sampled at few centimetres of the fault, do not recorded Sr isotopic evidence ($^{87}$Sr/$^{86}$Sr = 0.70929) of water-rock interaction. Nevertheless, Sr isotopic compositions of a slickenside sample ($^{87}$Sr/$^{86}$Sr = 0.70886) and a MSL clast of the fault breccia ($^{87}$Sr/$^{86}$Sr = 0.70893) suggest a slight interaction with the paleofluid circulating along the fault.

3/ Pb isotopic composition of authigenic or early diagenetic pyrite spheroids are less radiogenic (18,628 $\leq$ $^{206}$Pb/$^{204}$Pb $\leq$ 18,679) than those of cubic pyrites (18,736 $\leq$ $^{206}$Pb/$^{204}$Pb $\leq$ 18,749) filling with calcite, tension gashes in the vicinity of the studied fault zone. In the Pb-Pb isochron diagram, Pb isotope evolution of pyrites suggests a crystallization age of 86 Ma for the authigenic pyrites disseminated in the MSL and a 39 Ma age for the fluid paleocirculation in the fault, i.e. at the end of the Pyrenean compressionnal event or at the beginning of the Oligocene rifting.

4/ Marcoule silty-clays from the MAR 202 and 203 boreholes do not show the Sr and Pb isotopic anomalies recorded in the vicinity of the MAR 501 fault zone. Rb/Sr, U/ Pb and Pb/Pb data are consistent with an evolution of the isotopic tracers in closed system conditions since 100 Ma, without exchange of radiogenic or radioactive isotopes by water-rock interaction, in spite of the onset of the Pyrenean and Alpine orogenies, the Oligocene rifting and the Messinian event.
References


Figure 1a: Rb/Sr isochron diagram for cogenetic and synchronous rocks of magmatic or metamorphic origin, evolving in closed system with the time. The $\alpha$ slope of the whole-rock isochron corresponds to $t$, which is the time elapsed since crystallization of the rocks and the y-intercept yields their Sr initial ratio ($^{87}\text{Sr}/^{86}\text{Sr})_0$. The slope provides $t$, age of crystallization. (□) initial location of the points; (■) location of the points at present time.
Figure 1b: Rb/Sr isochron diagram for cogenetic and synchronous sedimentary rocks. The Rb/Sr evolution in closed system with the time is controlled by the mixing of C, an unradiogenic end-member (diagenetic carbonates), with M, a radiogenic end-member ones (detrital muscovites and clays). The y-intercept of the mixing line yields the Sr ratio of diagenetic carbonates. The $\alpha'$ slope provides a $t'$ age greater than $t$, time of deposition of the sediments. (□) initial location of the points; (■) location of the points at present time; (●) C unradiogenic end-member; M radiogenic end-member, (△) at $t$, (▲) at present time.
Figure 2: Sketch geological map of the studied area after [4] and stratigraphic correlations between MAR 501 and 203 boreholes after [5].
Fig. 3: Rb/Sr and U/Pb mixing lines for MSL whole rocks in isochron diagrams. The data points represent MSL samples from MAR 202-203 and MAR 501 boreholes (O, ●), three Gargasian marls sampled on the field (□, ■), acid-leached Gargasian carbonates (◇), MSL disseminated pyrites () and Gargasian disseminated pyrites (). Y-intercept yields the $^{87}$Sr/$^{86}$Sr ratio of MSL diagenetic carbonates. In each isochron diagram, the 100 Ma isochron shown as reference is calculated for initial $^{87}$Sr/$^{86}$Sr and $^{206}$Pb/$^{204}$Pb values respectively of 0.70748 and 18.65.
Figure 4: Evolution of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios with time in MSL (in dark grey), overlying series (in grey) and underlying series (in light grey) rocks from MAR 202 and 203 boreholes. The Rb/Sr evolution field of MSL is obviously smaller than those of the overlying (Cenomanian) and underlying (Aptian) formations where aquifers are located at present time.
Figure 5: Depth profiles of $^{87}\text{Sr}/^{86}\text{Sr}$, $^{87}\text{Rb}/^{86}\text{Sr}$, $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{238}\text{U}/^{204}\text{Pb}$ ratios in MAR 202, 203 and 501 boreholes. These diagrams outline the homogeneity of MSL in comparison with the heterogeneity of Cenomanian and Aptian formations, related both to petrological variations in sediments overlying or underlying the MSL and to water-rock interactions in upper and lower aquifers. (O) present-day isotopic values in MAR 202 and 203 boreholes; (●) present-day isotopic values in MAR 501 borehole; (●) initial isotopic values in MAR 202 and 203 boreholes.
Figure 6a: Variation of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of MSL whole rocks and mineral fractions (calcite, celestite) sampled in a fault or in its vicinity (MAR 501 borehole).

Figure 6b: Variation of $^{206}\text{Pb}/^{204}\text{Pb}$ ratio of pyrites in the vicinity of the studied fault: (●) disseminated authigenic or early diagenetic pyrite in MSL sediments; in comparison, disseminated pyrites in the MSL from the MAR 202 borehole and in Gargasian marls sampled on the field, provide similar $^{206}\text{Pb}/^{204}\text{Pb}$ ratios respectively of $18.659 \pm 0.004$ and $18.660 \pm 0.001$; (○) cubic pyrites crystallized (with calcite and celestite) both in tension gashes located on each side of the fault and in the cement of the fault breccia.

See pp. 238-239.
Minerals sampled in or along the fault
Ca: Calcite, Ce: Celestite

87Sr/86Sr

0.70973 ± 0.00003

0.70970 ± 0.00002

0.70913 ± 0.00002

0.70933 ± 0.00003

0.70919 ± 0.00002

0.70961 ± 0.00001

0.70911 ± 0.00003

0.70929 ± 2 (WR-1)

0.70824 ± 2 (WR-2)

0.7086 ± 2 (WR-5)

0.7084 ± 1 (WR-4)

0.70893 ± 1 (WR-3)

WR: Whole rocks sampled in or along the fault
WR-1: without calcite gashes
WR-2 and WR-4: with calcite gashes
WR-3: MS& class in fault breccia
WR-5: slickenside sample
Figure 7: Variation of the $^{87}$Sr/$^{86}$Sr ratio of MSL whole rocks in the vicinity of a fault (MAR 501 borehole); (O) whole rocks with tension gashes; (●) whole rocks without tension gashes; (■) calcite and celestite filling tension gashes and the cement of the fault breccia. The isotope composition of Sr recorded by diagenetic carbonates ($0.70741 \leq ^{87}$Sr/$^{86}$Sr $\leq 0.70758$) is very similar to those of calcites and celestites ($0.70749 \leq ^{87}$Sr/$^{86}$Sr $\leq 0.70768$).
Fig. 8: Isotopic compositions of Pb in pyrites from MAR 202-203 and 501 boreholes; (●) MSL disseminated pyrites; (○) disseminated pyrites in Gargasan marls; (□) average of disseminated pyrites; (■) cubic pyrites from the fault breccia and the tension gashes; (□) average of cubic pyrites. Two-stage growth curves (μ₂ egal to 9.74, 9.81 and 10) and secondary isochrons of the Stacey-Kramers model [22] are shown for reference. Pb isotope evolution of pyrites suggests a 42 Ma (Ş.-K. model) age for the fluid flow in the MSL through the studied fault of the MAR 501 borehole.
Characterization of Gas Flow in Boom Clay, a Low Permeability Plastic Rock

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Abstract

Within the MEGAS project, experiments and modelling on gas migration through a Boom clay host rock have been undertaken. The main purpose of this research was to assess the global influence of gas generation and migration on the safety of an underground radioactive waste repository, located in the middle of the Boom Clay layer, located under the CEN•SCK site in Mol, Belgium. A key aspect of the MEGAS project is its multi-disciplinary approach, with experiments undertaken by CEN•SCK, BGS and ISMES and designed in collaboration with the modelling team at QuantiSci. The project has been co-funded by the European Commission and NIRAS/ONDRAF.

A series of gas injection experiments on small and large cylindrical clay cores were undertaken in the laboratory. The experimental procedure consisted of three different phases: the clay core was first completely resaturated, then its hydraulic conductivity was measured, and finally gas injection was started. During this latter phase, the gas injection pressure was increased step wise until a breakthrough was observed at the outlet of the sample.

We made several important observations, that were repeatable throughout these laboratory experiments:

- Gas breakthrough is associated with the creation of preferential pathways, and occurs at pressures close to zero effective stress conditions. Gas seems to move in "bursts" along these pathways, the aperture and closure of the pathway depending on local geomechanical stress conditions. A pathway will open or close if a given threshold is crossed by the gas pressure.
- A non-Darcy behaviour of gas flow rate versus gas pressure was observed. Indeed, a strong increase of gas flow resulted from small changes in pressure.
- No major desaturation resulted from the breakthrough, with 2% being the total desaturation measured for the three cores.
- A correlation seems to exist between the breakthrough pressure and the hydraulic conductivity [1] and [2]. Both parameters varied in a wide range for the complete set of samples. We measured hydraulic conductivities ranging from 1x10^{-12} to 7x10^{-12} m.s^{-1} and breakthrough pressure between 1 and 3 MPa.
The major role of the effective stress in the gas migration process was also established during two in situ gas injection experiments, performed in two distinct areas of the Boom Clay layer. In these experiments, gas was injected through one of the screens of a multipiezometer installed in the clay layer from the HADES underground laboratory. A displacement transducer measured the gas inflow, and a pressure transducer connected to each of the neighbour screens measured the local interstitial pressure. At the breakthrough, we observed a sudden formation of a preferential pathway along the injection multipiezometer, which is obviously a path of least resistance. This phenomenon happened at a breakthrough pressure which was quite inferior to the values measured in laboratory on clay samples, and can be explained by a local drop of the effective stress around the borehole [3], subsequent to the clay convergence occurred during the drilling.

Attempts to model the gas migration behaviour using a traditional Darcy two-phase flow model were successful in predicting steady-state gas fluxes, but any match to the experimentally observed transient behaviour could only be obtained by making the porosity and relative permeabilities functions of the effective stress. Further progress was made by applying a recently developed capillary bundle model, which directly represents the preferential pathways for gas by a series of capillary tubes. The capillary bundle model successfully predicted both the transient and steady-state behaviour observed in experiments.

MEGAS will be continued within the PROGRESS project, which will focus on the development of the understanding of the formation of preferential pathways in Boom clay. On the experimental side, more gas injection experiments are foreseen on large clay cores in isostatic cells, with special attention to the detection of preferential pathways. To test the influence of a destressed zone around excavations on the gas breakthrough pressure and the gas flow, experiments will be performed at different total and effective stress states. The long term effects of gas flow on material performance, and the self healing capacity of the Boom Clay after the formation of preferential pathways will also be investigated, in laboratory and in situ. Another objective of the experimental work will be to measure the source term for the gas generation rate in real conditions.

In terms of modelling, attention will be focused on extending the capillary bundle model to incorporate realistic geomechanical effects. Initial work has considered the incorporation of generic "pathway formation" and "pathway closure" models, in which pathways are dynamically formed by the displacement of clay platelets, and pathways close to gas flow if the gas pressure drops below a threshold value related to the effective stress. These simple models exhibit realistic behaviour including "bursting", and also illustrate how the capillary bundle model provides an ideal framework in which further physical and chemical effects can be incorporated.
1. Introduction

In a deep geological radioactive waste repository, hydrogen gas may be generated in significant quantities. It will be produced mainly by the anaerobic corrosion of the metallic waste canisters. This gas production may induce a pressure build-up around the repository. The two main questions concerning the safety are: how will the gas escape in the rock medium and what will be the influence of this pressure build-up on the rock properties? Several phenomena that could lead to a pressure decrease have been faced and assessed quantitatively during MEGAS: the consumption of hydrogen by a chemical reaction with Boom clay, a diffusive flow of hydrogen into the clay massif, and an advective flow provoked by a gradient in hydraulic pressure, which would obey the Darcy's law, according to the first hypothesis considered in this research project. In this paper, we will not elaborate on the two first mechanisms, as it appeared that their respective contribution to the hydrogen consumption rate should be several orders of magnitude lower than the one due to Darcian flow.

Previous modelling of the MEGAS gas injection experiments utilized the TOPAZ two-phase flow model [4] which extended the classical Darcy two-phase flow model by allowing the permeability and porosity of the core to vary with gas pressure. This development was motivated by the fact that the classical Darcy model, based on a representative elementary volume (REV) or effective continuum approach, could predict the steady-state behaviour in some experiments but not the transient behaviour. It was demonstrated that pressure-dependent properties influenced the transient behaviour without significantly perturbing the steady-state behaviour, and thus that good fits to experimental data could be obtained. One interpretation of these results, that was also supported by experimental observations and heuristic arguments, was that preferential pathways could play a significant role in gas migration in Boom clay. The rationale for using an REV model in such a gas migration regime is currently the subject of much debate, and available alternative models are considered.

The results of an investigation of one possible alternative are presented. The approach, a capillary bundle model, is based on an idealized, topological representation of gas/water migration pathways through a Boom clay sample. The first objective is to demonstrate whether the capillary bundle model produces transient and steady-state behaviour consistent with MEGAS experimental results. Since the capillary bundle model has a large number of degrees of freedom, particular care is taken to avoid the investigation becoming a "curve-fitting exercise". The approach taken is to conduct simple scoping calculations of the MEGAS gas injection experiments using, as far as possible, generic information and simple experimental measurables.

A second objective is to consider whether the capillary bundle model can provide support for the main conclusion arrived at in the MEGAS project, namely that preferential pathways play a significant role in gas migration in Boom clay. The reasoning here is that the capillary bundle model employs a much more direct (i.e. physically understandable) representation of preferential pathways than the current REV models and so may be a better tool with which to decide whether preferential pathways are indeed as significant as thought. This does not preclude further development of the REV approach, since the physical insight gained by use of the capillary bundle model could lead to a better understanding of how the more orthodox REV formulation (with all its historical advantages of data sources and availability) could be modified to take account of preferential pathways.
2. Laboratory gas breakthrough experiments

2.1. On small Boom Clay cores in a permeameter cell

The basic experimental set-up for the gas breakthrough experiments is shown in Fig. 1, and is essentially composed of three parts: a permeameter cell and two flow monitoring systems. The detection limit of the flow monitoring systems, represented by a displacement transducer, is in the range of a microliter per minute.

![Diagram of experimental set-up for breakthrough experiments](image)

Figure 1. Experimental set-up for breakthrough experiments

The internal geometry of the permeameter cell allowed us to test samples with 38 mm diameter and up to 100 mm long. To allow different types of experiments, three different versions of this basic set-up have been built [5]:

- To realize experiments on fully constrained samples, the permeameter cell is placed in an adaptable rigid frame, and the position of the cell's piston is blocked by a screw.

- We placed the permeameter cell under a mechanical loadframe to carry out experiments on samples placed under a constant vertical load of 4.4 MPa. It simulates the total in situ stress that prevails in the Boom Clay layer at the depth of the HADES underground research facility. We used a displacement gauge with a precision of 2 μm to measure the consolidation ratio of the clay sample.

- We used also X-ray transparent plexiglass cells to be able to measure continuously the saturation profile of the clay samples during the gas breakthrough, by taking tomographies of the cell.

We applied the following general procedure to carry out the gas breakthrough experiments:

We measure all the physical parameters of the clay sample (length, density, weight, porosity). After the installation of the clay sample in the permeameter cell, the resaturation phase begins. Water is injected at the bottom side of the plug, until the in- and outflow equilibrate. When the sample is fully resaturated, we calculate its hydraulic conductivity by applying Darcy's law. Then, for the corresponding
experiments, we apply the vertical load, and we wait for the reconsolidation of the sample. When it is in hydro-mechanical equilibrium, we measure again all the physical parameters, and the hydraulic conductivity of the sample. During all the experimental process, we keep the water pressure $P_w$ at the bottom of the clay sample at a constant value of 2.2 MPa, which represents the water column at the depth of the HADES facility. Helium is then injected at the top of the sample, at constant pressure conditions $P_g = 2.3$ MPa. When we don't observe any more water production at the outlet of the sample, we increase $P_g$ in steps of 0.1 MPa, until a gas breakthrough is established. The gas outflow is monitored, until the upper limit of detection of the displacement transducer is reached. We measured also the saturation profile of the clay sample: non-destructively during all the gas injection process for the X-ray transparent cells, destructively after the breakthrough occurrence for all the other experiments.

Figures 2 and 3 show typical examples of two distinct evolutions observed of the gas flow-rate at breakthrough. In sample 23B7.5K2 (Fig. 2), the gas flow sharply increased after breakthrough, while in the case of sample 23B8.5K2 (Fig. 3), the gas flow increases gradually during several days, then stabilizes. The first behaviour was more frequent and is due to the creation of preferential pathways, while the second behaviour is closer to what could be expected on the basis of an extended Darcy flow type of model, and was only observed once during the course of the gas breakthrough experiments on small clay cores.

![Flow rate vs. Time](image1.png)

**Figure 2. Example of gas migration by the formation of preferential pathways in sample 23B7.5K2**

![Flow rate vs. Time](image2.png)

**Figure 3. Example of a gas migration mechanism assimilated as two-phase flow in sample 23B8.5K2**

### 2.2 On large clay cores in isostatic cells

A large dispersivity in the results was observed during the gas breakthrough experiments realized with small clay cores. So, it was decided to carry out gas injection experiments on larger cylindrical clay cores (length: 300 mm, diameter: 80 mm) to reduce the influence of the scale effect and the inhomogeneities on the experimental results. We used therefore three home-built isostatic cells, in which it is possible to control precisely the isostatic confinement pressure and the effective stress in the sample. We carried out three experiments at the same time.

The isostatic cells are composed of three main parts: a cylindrical body, a hemispherical top and a flat base. The cells have been conceived to work under a nominal pressure of 4.5 MPa. Two stainless steel caps, carrying a sintered polyethylene filter are placed in contact with the extremities of the clay sample. This sample is first wrapped in an aluminized polyethylene foil to avoid later the propagation of the
injected gas by diffusion along the walls. The whole assembly (steels caps, clay sample and aluminized foil) is slipped into a neoprene membrane, and screwed together with the pipe-work leading. The three parts of the isostatic cell are then assembled. Each isostatic cell is connected to a common circuit used to apply the confining pressure on the samples. We chose water as confining fluid. Temperature controlled water at 30°C is pumped through a plastic coil, wrapped around the cylindrical body of the cells to avoid large fluctuations of the confining pressure. A temperature probe was placed in each cell to measure the fluctuations of temperature.

When the isostatic cells were full with water, a confinement pressure of 4.3 MPa was applied. Immediately afterwards, the resaturation process of the clay samples was started. Water was injected through the sample bottom. A pore water pressure was imposed upstream and downstream the sample. The pressure gradient along the core was chosen to be as small as possible to keep an uniform stress field along the sample, but large enough though to have a reasonable and measurable water flow, to resaturate the sample as fast as convenient. The average pore water pressure was set at 2.2 MPa.

For cell no. 1, two syringe pumps with an internal volume of 100 ml, are connected to each face of the Boom Clay sample. This very accurate apparatus allows us to work either at constant pressure or at constant flow. Flow-rates as low as 10⁻⁵ mL.min⁻¹ can be measured or imposed. The syringe pump located downstream the sample imposed a constant backpressure. Upstream the sample, the other syringe pump works in constant pressure mode during the saturation process. Then, when the injection of helium started, the syringe pump was switched to constant flow mode in cell no.1. We chose to apply a constant pressure boundary condition at the inlet of the two other samples. The data provided by the syringe pumps are recorded every ninety seconds by a data acquisition system.

The experimental histories of the three core samples are quite similar. Up to a gas pressure of 4.2 MPa, no breakthrough was observed. When the inlet pressure reached 4.2 MPa in cell no.1, we switched the boundary conditions from constant flow to constant pressure, and we continued the experiment by decreasing step wise the downstream water pressure until a breakthrough was detected. After seven months of experiments, the respective backpressures at the outlet of the samples were equal to 1.4 MPa in cell no.1 and 1.5 MPa in the two other cells. A breakdown of the motor of the water temperature regulator caused a drop of the confining fluid temperature from 25 to 21°C, and provoked a decrease of the confining pressure in the three cells. In cells no. 2 and 3, the inlet gas pressure became larger than the confining pressure during a few hours. This situation resulted in a breakthrough in the three samples, obviously caused by the relaxation of the isostatic stress in the clay samples. Some very important observations support the hypothesis of the determining role of the effective stress in the gas breakthrough preparation:

- In cell no. 1, the gas pressure has always been lower than the confining pressure. That excludes the possibility of a gas flow along the walls of the clay core. Moreover, it shows that the breakthrough occur when the total stress is still about 0.02 MPa larger than the gas pressure.

- In the two other cells, the breakthrough occurred several hours after the beginning of the isostatic stress decrease [6]. A possible explanation is that the drop of the effective stress is a delayed process (at first, it is the interstitial pore pressure that follows the evolution of the total isostatic stress)

The following features were observed for the three samples:

- A measurable quantity of gas penetrated the clay core several hours before the detection of the breakthrough at the other extremity of the clay sample.
A previous desaturation of about 2% was measured before the breakthrough occurrence.

The breakthrough was announced in every sample by a preliminary preferential pathway opening, which time for formation was quite repeatable and lasted about fifteen minutes in every sample.

After this preliminary pathway opening, a “burst type” gas transport phenomenon has been observed in every sample. The frequency of these pathways opening and closure was inferior to the scanning period of the data acquisition system (which is ninety seconds). The peak flow amplitude increased notably by a factor three compared to the flow measured during the preliminary pathway opening phase.

There exist however some differences in the evolution of the gas transport process in the three samples:

- In system 1 and 3: a linear evolution of the expelled quantity of gas was observed during the breakthrough, until the gas flow was stopped by the subsequent pressure increase at the outlet of the sample due to the complete filling of the internal volume of the pump by the gas, and the equilibration of the pressures at the inlet and at the outlet of the sample.

- In system 2, the average flow decreased and evolved progressively to an equilibrium. The “burst type” flow ceased after 45 minutes and was replaced with a “smooth” flow evolution, which lasted for three more hours until the outlet compartment was filled with the gas at the imposed pressure of 1.5 MPa. Then, a slight pressure increase was observed at the outlet, to reach the equilibrium pressure of 1.7 MPa.

Figures 4 and 5 show the gas flow-rate evolution after the gas breakthrough in systems 1 and 2.

![Figure 4. Gas flow-rate evolution after gas breakthrough in system 1](image1)

![Figure 5. Gas flow-rate evolution after gas breakthrough in system 2](image2)

3. In situ experiments

In situ experiments have been performed at two different locations in the HADES underground research facility i.e. under the bottom of the main access shaft and at ring 20 in the “TEST DRIFT”. The experiments performed at these locations will be further referred to as respectively experiment E4 and experiment E5 and are described by Fig. 6 and 7.
3.1. The E4 gas injection experiment

In situ gas injection experiments using Helium were performed in a vertical piezonest. This piezometer has only one connection tube to each filter. Previous to gas injection in a screen the water contained in the connection tube and the dead volume of the filter has to be expelled by pushing it into the clay massif. This problem is avoided with the new MEGAS in situ set-up (E5). We made two subsequent gas injection experiments, in filters 6 and 9.
After the stabilization of the interstitial pressure around the piezometer, gas was injected through the filter no. 6. Gas was injected at such a pressure that it could set the filter free of water (up to 1.24 MPa), and be less than the threshold pressure for gas flow. Without taking into account the modification of the geomechanical properties due to the drilling, and knowing that filter no. 6 is about 13 meters below the tunnel axis level, the local total stress value was estimated to be between 2.5 and 4 MPa [7]. The pressure of 2.5 MPa was taken as the upper limit for the gas injection pressure in order to avoid any fracturing of the clay. The initial value for the gas pressure was chosen at 2.25 MPa, i.e. between the local estimated total stress and the interstitial pressure.

As well in the gas injection test performed on filter 6 as the injection test performed on filter 9, we obtained gas breakthrough at much lower gas overpressures than expected i.e. about 0.65 MPa instead of 1.3 MPa and 1.7 MPa, which are the local effective stress estimates. In the case of the injection test through filter 9, breakthrough occurred over the full length of the piezometer upwards from filter 9 and the experiment was stopped, because gas was entering the shaft.

The most important phenomena observed are the following: the breakthrough pressure was quite lower than expected and one or more preferential pathways were created in the direction upwards to the injection filter and along the piezometer. This can be explained by the influence of the drilling of the borehole on the local geomechanical stress distribution [3].

3.2. The E5 gas injection experiment

The new in situ gas injection experiment E5 consists in four horizontally installed piezometers. The central piezometer (90 mm diameter) is used for the gas injection while the three peripheral 60 mm diameter piezometers are used for the detection of pressure changes caused by gas or water injection. Two detection piezometers are installed in the horizontal plane at different distances from the injection filter. In the vertical plane only one detection piezometer has been installed. This configuration was chosen because the breakthrough experiments have shown that due to the anisotropy in the hydraulic conductivity it can be expected that breakthrough will occur more easily in the horizontal direction than in the vertical direction. In total there are 29 filters, which are each connected to a pressure transducer. Each filter is equipped with two connection tubes (int. φ 2 mm, ext. φ 4 mm). This allows us to conveniently purge the filter.

![Diagram of pressure evolution in filters 18 to 21 during the gas injection experiment](image)

Figure 8. Pressure evolution in filters 18 to 21 during the gas injection experiment

After the hydrostatic pressures of the piezometers had reached equilibrium, helium was injected in filter 20. The gas injection pressure was first set at 0.1 MPa above the local pore water pressure, which was equal to 1.67 MPa. Then, the gas pressure was increased weekly breakthrough was reached. by increments 0.1 MPa until gas breakthrough to filter 19 occurred after 44 days at a gas pressure of 2.36 MPa, i.e. 0.69 MPa above the original pore water pressure. The pressures
in filter 20 and 19 became almost equal (see Fig. 8). This shows that between those filters a preferential pathway was created by the gas. A pressure increase on the order of 0.2 MPa was detected in several peripheral filters at breakthrough time. This is a strong indication that the creation of a preferential pathway is due to a hydromechanical effect.

The first 12 days after gas breakthrough the gas flow rate was relatively high (i.e. 0.3 ml STP.min⁻¹). The gas probably pushed the water in the collection chamber of filter 19 into the clay. Later the gas pressure was increased to 2.43 MPa and a constant gas flow rate of 0.115 ml STP.min⁻¹ was observed. This gas flux is of the same order of magnitude as the gas flux measured in the laboratory experiments.

When the gas injection pressure was further increased to 2.53 MPa breakthrough occurred on filter 18 and one day later also on filter 21 both situated on the injection piezometer. The pressure on all these filters became almost equal, indicating a direct connection between them. These results confirm the observations of the E4 experiment i.e. the in situ gas flow behaviour is governed by the coupling between the geomechanical and the hydraulic behaviour of the Boom clay.

4. Modelling

4.1. The capillary bundle model

In the capillary bundle model we assume that the core is composed of a series of non-intersecting capillaries [8]. This is an idealised topological representation of the gas/water transport pathways in the core, as shown in Fig. 9.

We assume that each capillary has uniform radius \( r \); and that the number of capillaries per unit cross sectional area of core, with radius in the range \([r, r+dr]\), is \(N(r)dr\). The length on average of each capillary is \(\tau L\). \(\tau\) is known as the tortuosity factor. The combined volume of all the capillaries must be equal to the volume available for fluid motion. Hence:

\[
\int_0^\infty \tau L \cdot \pi r^2 \cdot N(r) \ A \ dr = \Theta \cdot A \cdot L
\]

We can now introduce a more convenient distribution, that is the cross sectional area per radius per unit cross sectional area of core. It is given by

\[
g(r) = \pi r^2 \cdot N(r) .
\]

Following [9], if the length of the portion of a capillary of radius \( r \) (measured along the capillary) which is filled with water is \( R(r,t) \), then assuming slow flow within the water filled portion of the capillary, by
solving the Navier-Stokes equation [10] the position \( R(r, t) \) of the gas-water interface can be shown to be:

\[
\frac{dR(r, t)}{dt} = \frac{1}{R(r, t)} \left( r \sqrt{2\sigma} + r^2 \frac{P_g - P_w}{8\mu_w} \right)
\]

(3)

where \( P_w, P_g \) and \( P_c \) are the water, gas and capillary pressures respectively and \( \mu_w \) is the dynamic viscosity of water. The capillary pressure is given by

\[
P_c = \frac{2\sigma \cos \alpha}{r}
\]

(4)

where \( \sigma \) is the surface tension and \( \alpha \) is the angle of contact (\( = \pi/4 \)). Hence if the gas and water pressures are constant, and the capillary is initially totally full of water, that is \( R(r, 0) = \pi L \), then:

\[
R^2(r, t) = \pi^2 L^2 - \frac{t}{4\mu_w} \left( r^2 (P_g - P_w) - r \sqrt{2\sigma} \right)
\]

(5)

which is valid in the region \( 0 \leq R(r, t) \leq \pi L \).

The gas pressure must exceed the water and capillary pressure combined in order for a capillary to start desaturating. The smallest capillary radius, \( r_s \), which starts desaturating at given gas and water pressure, is given by

\[
r_s = \frac{\sqrt{2}\sigma}{(P_g - P_w)}
\]

(6)

Capillaries with radius smaller than \( r_s \) will remain totally saturated. At fixed gas and water pressure each capillary with radius greater than \( r_s \) will desaturate until gas breakthrough, that is when the length of the water filled part is zero. Hence at time \( t \) the largest capillary which contributes to the total flux of water from the capillaries has radius

\[
r_f(t) = \sqrt{\frac{\sigma + \sqrt{\sigma^2 + \frac{8\mu_w \pi^2 L^2 (P_g - P_w)}}}{t} \left( P_g - P_w \right)}
\]

(7)

Hence the total flux of water from the core is given by:

\[
\Phi_w(t) = \int_{r_f}^{r_s} \Phi_w(r, t) N(r) A \, dr
\]

(8)

After breakthrough in a capillary of radius \( r \) at time \( t_b(r) \), the gas pressure distribution along the capillary reaches equilibrium at a rate controlled by conservation of mass and Darcy's Law. The saturation profile

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of the core can be calculated at any point by summing the contribution of each capillary. At a point \( x \) along the core, the capillaries which contribute, must have \( R(r, t) \propto x \). Larger capillaries will be emptied beyond this limit. Hence at time \( t \) the largest capillary which contributes at the distance \( x \) along the core is

\[
R_x(x, t) = \frac{\sigma}{\sqrt{2 \pi}} \frac{8 \mu}{\eta} \tau^2 \left( \frac{L^2 - x^2}{t} \right) \left( \frac{P_x - P_w}{\sqrt{2 (P_g - P_w)}} \right)
\]

(9)

The saturation \( s(x, t) \) along the core is given by

\[
s(x, t) = \int_0^{r_x(x, t)} g(r) \, dr \cdot \left( \int_0^{r_x(x, t)} g(r) \, dr \right)^{-1}
\]

(10)

4.2. Results

The following figure 10 shows typical results from the comparison of the capillary bundle model with experimental data and TOPAZ calculated values. The values of the variables and distribution used are given in [11]

![Figure 10. Total gas flux plotted as a function of time for core 23B8.5K2](image)

The main conclusion of the modelling is that the steady-state and transient behaviour of the capillary bundle model is in good agreement with the MEGAS experimental results. This provides support for the hypothesis that preferential pathways play a significant role in gas migration through Boom clay. In
reality, the method of gas transport may be a combination of two processes, that is two phase flow and interfacial evolution within preferential pathways, but it is significant that by using simple but realistic models for the pore radii distribution, one may produce results which show both qualitative and quantitative features of the experimental results. Were wetting to be also considered such models also exhibit natural hysteresis [9].

Extensions of the capillary bundle model which should allow further examination of gas migration along preferential pathways include:

- allowing the pore radii to be function of the pore length, gas pressure and time
- allowing connections between capillaries [12]

A significant issue that remains to be addressed is the use of model parameters such as $\varepsilon$, $D_g$ and $g(r)$, which may be difficult to measure experimentally. This issue is particularly important for the pore radii distribution function, $g(r)$, since it introduces degrees of freedom which could be exploited to "curve-fit" model behaviour to experimental behaviour. Care has been taken here to avoid this by using a generic function. Indeed, the number of degrees of freedom used is equivalent to those used in two-phase Darcy models. The success of the capillary bundle reported here indicates that further investigation of these issues and detailed consideration of a modelling/experimental study is certainly worthwhile.

5. Conclusions

Our experimental and modelling work performed during the MEGAS project has clearly put in evidence the predominant role played by the hydromechanical coupling in the gas transport mechanism through a low permeability plastic clay. This assertion has been proved further by the results obtained from a hydraulic test performed in the vicinity of the E5 installation [13].

In a high-level radioactive repository, there will be a gas pressure build-up generated by the gas production, mainly induced by the anaerobic corrosion of the metallic package of the waste canisters and the construction material. This will eventually lead to a breakthrough when the local zero effective conditions will be reached. Gas will flow through preferential pathway(s), which will follow the path of least resistance in the massif, i.e. the spatial direction where the tensor component of the effective stress will be the lowest.

Previous modelling work carried out during the MEGAS project tend to prove that the capillary bundle approach is promising. A more sophisticated version of the model, that will implement the stress dependence of the pore radii distribution and the possibility of connections between the paths is though required.

Further investigations are needed to study deeper the phenomenology involved. These research topics will be included in two new projects, PROGRESS and RESEAL, which are part of the fourth framework programme of the CEC. These projects will focus on the following items:

- The elaboration of an hydromechanical model, that will describe the behaviour of the intact clay host rock matrix on the one hand, and of the backfilling and sealing material (composed in majority of calcic bentonite) on the other hand. Some laboratory experiments will provide the data set necessary for its validation;
- The self-healing capacity of these respective materials;
- The gas generation source term;
- The in situ installation procedure at large scale of a seal in an excavation.
6. Acknowledgements

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7. References


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POSTER SESSION
Irreversible Deformation and its Incidence on the Hydraulic Properties of Clays

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Abstract

Natural fluid overpressures occurring in fine grained sediments can be released by two different mechanisms: large scale natural hydrofracturing or smaller scale damages associated with dilatancy that enhance permeability. We focus on this second mechanism.

Three geological situations can lead to stress path pointing towards the dilatant part of the yield envelope of a compacting rock: first the hydraulic connection of the compacting rock to an overpressurised lower reservoir, second an erosion that affects the vertical stress magnitude, and third a change in the tectonic regime that reduces at least one of the horizontal stress magnitudes.

An experimental work is currently under way to define the conditions under which dilatancy sufficiently enhances permeability to release overpressures without requiring large fractures. Remoulded clays are first consolidated to high pressures (20 MPa) in an oedometric cell. They are then loaded in a triaxial apparatus where a stress path leading to dilatancy is applied and the permeability is monitored by measuring fluid flows. The evolution of the pore space is further analysed by X-ray tomography and microstructural observations.

The different behaviour of clays obtained in traction and compression experiments suggests to differentiate the yield envelope for each case.
INTRODUCTION

During the sedimentation of clays, if the porosity reduction due to compaction leads to a large decrease of permeability, pore fluids may become trapped and pore pressure may increase significantly above hydrostatic levels. Two mechanisms can release these fluid overpressures: large scale natural hydrofracturing or smaller scale damages associated with dilatancy that enhance permeability. We focus on this second mechanism and summarise the geophysical conditions it requires.

We then summarise the procedure and very preliminary results of an ongoing experimental work that aims to define the conditions under which dilatancy sufficiently enhances permeability to release overpressures without requiring large fractures. This work is integrated in and supported by the ‘Groupement de Recherche Géomécanique des Roches Profondes’ program. The material to be tested was to reveal the hydro-mechanical coupling as simply as possible. We chose remoulded clays in order to ensure a good reproducibility of the experiments and kaolinite because it only weakly interacts with water and is sufficiently permeable to allow reasonable test durations. The selected kaolinite from Saint-Austell has a rather homogeneous particle size of around 1 μm and was extensively characterised in former work [1-4].

We finally comment on the different behaviour of clays between compressional and extensional triaxial loading and the implication of this difference in terms of yield envelope.

<table>
<thead>
<tr>
<th>Symbols</th>
<th>Comments</th>
</tr>
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<tbody>
<tr>
<td>$\sigma_1 \geq \sigma_2 \geq \sigma_3$</td>
<td>Principal stress magnitudes; compressive stresses are positive (rock mechanics convention)</td>
</tr>
<tr>
<td>$\sigma_{h1} \geq \sigma_{h2}, \sigma_v$</td>
<td>Maximum horizontal, minimum horizontal and vertical principal stress magnitudes</td>
</tr>
<tr>
<td>$P_p$</td>
<td>Pore pressure</td>
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<td>$p = (\sigma_1 + \sigma_2 + \sigma_3) / 3$</td>
<td>Mean stress</td>
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<td>$q = \sigma_1 - \sigma_3$</td>
<td>Stress difference</td>
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<td>$p' = p - P_p$</td>
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</tr>
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<td>$\sigma_a$</td>
<td>Axial stress</td>
</tr>
<tr>
<td>$\sigma_r$</td>
<td>Radial stress</td>
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<td>$\varepsilon_1 \geq \varepsilon_2 \geq \varepsilon_3$</td>
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</tr>
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<td>$\varepsilon_p = \varepsilon_1 + \varepsilon_2 + \varepsilon_3$</td>
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<td>$ep \cdot hc$</td>
<td>Volumetric strain derived from axial displacement and confining fluid volume in triaxial tests</td>
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<tr>
<td>$ep \cdot f$</td>
<td>Volumetric strain derived from pore fluid volume in triaxial tests</td>
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<tr>
<td>$\varepsilon_a = 2(\varepsilon_1 - \varepsilon_3) / 3$</td>
<td>Distortion</td>
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<td>Distortion derived from axial displacement and confining fluid volume in triaxial tests</td>
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<td>Confining fluid injection volume</td>
</tr>
<tr>
<td>$p \cdot (P_{fs} + P_{fi}) / 2$</td>
<td>At hydraulic steady state: $p \cdot (P_{fs} + P_{fi}) / 2$</td>
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<td>$K_l = \sigma_l / \sigma_a$</td>
<td>Stress ratio during oedometric loading</td>
</tr>
<tr>
<td>$K_u = \Delta \sigma_l / \Delta \sigma_a$</td>
<td>Incremental stress ratio during oedometric elastic unloading</td>
</tr>
</tbody>
</table>

Table 1. Symbols convention
NATURAL STRESS PATHS

Typically, dilatant behaviour is obtained for stress path with an effective mean stress well below the consolidation effective mean stress [5]. In other words, to obtain dilatant behaviour the material first need have been overconsolidated.

Consolidation is naturally occurring during sedimentation where the vertical principal stress, $\sigma_v$ (see Table 1 for notation conventions), corresponds to the greatest principal stress, $\sigma_1$, i.e. the most compressive principal stress, and increases as burial proceeds. The sediments thus undergo normal consolidation (Fig. 1a) and, assuming oedometric boundary conditions (no lateral displacement), the horizontal, $\sigma_h1$ and $\sigma_h2$, and vertical, $\sigma_v$, principal stresses are related by:

$$\sigma_h1 - P_p = K_l (\sigma_v - P_p)$$
$$\sigma_h2 - P_p = K_l (\sigma_v - P_p)$$

where $P_p$ is the pore pressure. Thus the tectonic regime [6] is extensional and pore pressure simply reduces the effect of the increasing load due to burial but does not modify the regime.

![Fig. 1. Stress paths in the ($p'$, $q$) plane. (a) Oedometric consolidation or sedimentary burial (b) Pore pressure rise after sedimentation (c) Unloading (d) Superposition of an extensional tectonic regime (e) Superposition of a wrench tectonic regime (f) Triaxial tests](image_url)
To generate an effective stress path pointing towards the dilatant part of the envelope requires a deviation from this simple compaction process. Three mechanisms can be thought of: first the hydraulic connection of the compacting rock to an overpressurised lower reservoir, second an erosion that affects the vertical stress magnitude, and third a change in the tectonic regime that affects the horizontal stress magnitudes.

The hydraulic connection to an overpressurized lower reservoir would mainly reduce the mean effective stress, \( p' \), without affecting the stress difference, \( q \), and is thus a plausible mechanism to induce dilatancy (Fig. 1b); the rate at which the pore pressure would rise would be controlled by the initial permeability of the clay layer.

An erosion would reduce the vertical principal stress. The variation of the horizontal effective principal stresses, \( \Delta \sigma h' \) and \( \Delta \sigma v' \), as a function of the variation of the vertical effective principal stress, \( \Delta \sigma v' \), can be assumed to follow that observed in oedometric unloading:

\[
\Delta \sigma h' = K_h \Delta \sigma v'
\]

\[
\Delta \sigma v' = K_v \Delta \sigma v'
\]

but the unloading incremental stress ratio, \( K_h \), is generally much smaller than the loading coefficient, \( K_l \) [4]. This implies the unloading stress path (path AB, Fig. 1c) has a much steeper slope than the loading stress path (Fig. 1a). As a consequence the horizontal stress may become equal to or larger than the vertical principal stress. In that case the stress path points towards the dilatant part of the yield envelope (path BC, Fig. 1c). Furthermore, if the permeability is sufficiently low and the unloading sufficiently fast, the initial pore pressure may persist and become anomalous for the new depth. This would shift the effective stress path further towards low mean effective stress values.

A superimposed tectonic stress can affect the two horizontal principal stresses in five different manners: (1) it can raise both, (2) raise only one and not the other, (3) raise one and lower the other, (4) lower one and not affect the other, or (5) lower both. This correspond to a superimposed compressional regime in the first two cases, a superimposed wrench regime in the next case, and a superimposed extensional regime in the last two cases. The mean stress is always raised in case 1 and 2, always lowered in cases 4 and 5 and may be lowered or increased in case 3. Thus dilatancy can arise only in cases 4 or 5 (Fig. 1d) or 3 (Fig. 1e).

**EXPERIMENTAL STRESS PATHS AND PROCEDURE**

The requirement of using overconsolidated clays to obtain dilatant behaviour in triaxial tests led us to a two step approach: first a slurry is consolidated to high pressure (typically 20MPa axial stress) in an oedometric cell and the resulting sample is then submitted to triaxial test.

The oedometric loading stress path is that of Fig. 1a but during unloading a path such as that of Fig. 1c is followed. The plastic strain during unloading thus results in a weakening of the material and a shrunk yield envelope.

The triaxial stress path is designed so as to reach the dilatant part of the now shrunk yield envelope through an isotropic compression followed by an increase of the stress difference while the mean stress is hold constant (Fig. 1f).

In order to assess the geometry of the cracks or pore volume increase associated with dilatancy the samples are analysed by X-ray tomography before and after the triaxial tests. Further characterisation result from the microscopic analyses of sample thin sections made after the tests. Once the experimental protocol is mastered, it is planned to test samples in a triaxial cell that is transparent to X-rays and is inserted within an X-ray scanner in order to image the density evolution as deformation proceeds.

**OEDOMETRIC CONSOLIDATION**

We specifically built an oedometric cell that is sufficiently high (400 mm) to produce typical triaxial test size samples of consolidated clay (40 mm radius and 800 mm height) and that can apply a sufficiently high consolidation axial stress (40 MPa) to simulate sedimentary burial of the order of the km. This cell is integrated in a system that stabilises the axial stress and records the axial strain and applied stress (Fig. 2).
Typical displacements and axial stress recorded during oedometric consolidation are shown on Fig. 3. The initial slurry water content was of 148% (the Atterberg liquidity and plasticity limits are respectively 66% and 33% [2]), the initial and final sample height were 210 mm and 74 mm respectively. Most of the axial strain occurs during the two first axial pressure steps, that is below 0.7 MPa. Former work in an oedometric cell where radial stress could be measured indicate values of K1 and Ku of 0.64 and 0.18 respectively [2,4].

Fig. 2. Oedometric test system
Fig. 3. Oedometric consolidation curves. (a) Axial stress. (b) Axial displacement
TRIAXIAL TESTS

The triaxial tests apparatus allows to control the axial and confining stress as well as the upper and lower pore fluid pressure (Fig. 4). It also records the axial displacement and the confining fluid flux as well as the upper and lower pore fluid flux. Keeping a slight difference between the upper and lower pore fluid pressures allows to measure the permeability of the sample when a steady state flow is obtained.

Triaxial tests are being done currently so that only results of two preliminary tests, SA4 and SA6, are available. Test SA6 ended prematurely before reaching the desired stress difference but otherwise followed the correct stress path. The corresponding raw data as a function of time are shown on Fig. 5.

A mean effective stress well below half the oedometric consolidation mean effective stress is chosen as target and the stress path is broken into three parts. First the total applied stress is increased isotropically while both fluid pressures are also increased (point A, Fig. 1f, and hours 0 to 25, Fig. 5a). Then, when the fluid pressure is deemed high enough, it is held constant while the total applied stress continues to increase isotropically (path AB, Fig. 1f, and hours 25 to 120, Fig. 5a). Finally, when the target mean effective stress is reached, the axial and confining pressure are adjusted so as to maintain the mean effective stress constant and to increase the stress difference (path BC, Fig. 1f, and hours 120 to 350, Fig. 5a).

The axial displacement (Fig. 5b) and either the confining fluid volume or the pore fluid (Fig. 5c) volume can be combined to infer the sample volumetric strain, $\varepsilon_p$, and distortion, $\varepsilon_q$, (Fig. 6a). Even though these two methods yield different estimates ($\varepsilon_p$ versus $\varepsilon_q$, Table 1) the general trend is consistent. The confining volume is sensitive to confining pressure variations, temperature variations that can be related either to external change or adiabatic evolution, to the jacket compressibility and to possible air bubbles in the confining circuit. Correction for all of these factors is difficult and it is often preferred to calibrate the system with an elastic cylinder of known properties. Since this was not done here, the pore fluid measure is thought to be the more reliable of the two estimates.
Fig. 5. Triaxial test data from sample SA6. The variables are explained in Table 1.

The evolution of the volumetric strain with respect to the mean effective stress can be approached by plotting it against the pressure $p_1$ defined as:

$$p_1 = \frac{1}{3} (\sigma_a + 2 \sigma_f) - \frac{1}{2} (P_{fs} + P_{ff})$$

where $P_{fs}$ and $P_{ff}$ are the measured (and controlled) upper and lower pore fluid pressure. Thus when the pore fluid flow is steady state, $p_1$ is a good approximation of the mean effective stress within the sample. The volumetric evolution follows a succession of steps on such a plot (Fig. 6b). The steep part corresponds to the brief time during which the load is increased ($p_1$ is increasing), whereas the flat part corresponds to volumetric change occurring at constant external total stress ($p_1$ is constant). This strain can be related to an increasing effective stress, $p'$, that corresponds to the hydraulic relaxation. The excess pore pressure, that is the pore pressure above average between $P_{fs}$ and $P_{ff}$, is schematically represented by the hatched areas of Fig. 6b and the likely effective stress profile, $p'$, is that below that hatched area. The main constraint is that $p_1$ and
Fig. 6. Volumetric strain analysis of the triaxial test data from sample SA6.

p' do coincide after enough time has passed for hydraulic relaxation. The hydraulic time constant can be estimated to be less than 10 hours as can be seen in the deformation duration (Fig. 6a) and as will be explained below based on permeability and pore fluid volume flow arguments. The increasing stress difference induces barely any volumetric strain (Fig. 6c). In the SA4 test a slight dilatancy was observed for higher values of stress difference than those reached here.

The pore fluid flow can be computed from the injected volume shown on Fig. 5c. The resulting flow is shown together with the load evolution on Fig. 7. After each load increment both upper and lower flow are outward going but around 10 hours later on one flow is inward and the other outward going with similar rates: this shows that hydraulic steady state is reached and that the flow thus indicates the sample permeability. During the first mean stress stage
Fig. 7. Permeability analysis of the triaxial test data from sample SA6

Fig. 8. Permeability evolution with consolidation
Fig. 9. Radiological density of sample SA4. Higher densities are lighter. (a) Longitudinal slice before triaxial test (b) Longitudinal slice after triaxial test. (c) Schematic position of line AB (d) Density values along line AB before triaxial test (e) Density values along similar line AB after triaxial test.
(p = 0.25 MPa) the permeability is about 14 μDarcy and during the following stages it is halved to 7 μDarcy (also 7 × 10⁻⁹ m⁻² or an hydraulic conductivity of 7 × 10⁻¹¹ ms⁻¹). The permeability so derived is compared on Fig. 8 with that obtained in oedometric test on the same material [2]. The permeabilities are reported as a function of the oedometric consolidation axial stress in both cases and are grossly compatible. However, in the case of the triaxial test, the sample has been unloaded after consolidation and only partially reloaded in the triaxial cell. It is thus natural that the values obtained from these triaxial test are higher than those obtained from the oedometric tests at the same consolidation axial stress. Further test will try to measure the permeability variation associated with dilatancy.

MICROSTRUCTURAL AND PORE GEOMETRY ANALYSIS

Another measurement of strain is done by comparing X-rays densitometry of the samples before and after triaxial test. The results of such measurements are show on Fig. 9 for sample SA4 that was loaded to rupture and displayed some dilatancy just before it. The initial sample is slightly heterogeneous (Fig. 9a). The final sample is more homogeneous and slightly less dense (Fig. 9b). Longitudinal cross sections (position indicated on Fig. 9c) of radiological density before and after the triaxial test are shown in Fig. 9d & 9e. The bulk density, ρ, is related to the radiological density units used here, Ru, by:

$$\rho = \frac{R_u}{1024}$$

This allows to evaluate the initial porosity variability to less than 6% and the average final porosity increase to around 3%. Part of the final porosity increase corresponds to the dilatancy.

BEHAVIOUR DOMAINS

The behaviour of clays is different when they are submitted to either tensional ($\sigma_3 < \sigma_2 < \sigma_1$) or compressional ($\sigma_3 = \sigma_2 < \sigma_1$) triaxial stress paths. There is barely any peak of strength in extension tests whereas such a peak is generally observed in compression tests when the mean stress remains below a threshold that is a characteristic of the material. However extension tests do damage the sample but, because the decrease of the axial stress also decreases the mean stress, the volume change, $\varepsilon_v$, always corresponds to dilatancy and the damage can only be seen when that dilatancy begins to increase more significantly. In the $(p', \varepsilon_p)$ plane this can be observed when the curve slope changes.

To take into account these different behaviours, the yield criterion should include not only the two first stress tensor invariant, but also the third invariant. We propose to use for that purpose the parameter, $J_m$, derived from the Lode angle as follows.

From the deviatoric stress tensor, $\mathbf{s}$, related to the total, $\sigma$, and effective, $\sigma'$, stress tensors by:

$$\mathbf{s} = \sigma \mathbf{I} \left[ \frac{\sigma_1 + \sigma_2 + \sigma_3}{3} \right] - \sigma' \mathbf{I}$$

where $\mathbf{I}$ is the identity matrix, are defined the second, $J_2$, and third, $J_3$, invariant of the deviatoric stress:

$$J_2 = \frac{1}{2} \text{tr}(\mathbf{s}^2)$$

$$J_3 = \frac{1}{3} \text{tr}(\mathbf{s}^3)$$

In triaxial tests where either $\sigma_2 = \sigma_1$ or $\sigma_2 = \sigma_3$, the stress difference, q, is related to the second invariant by:

$$q = \sqrt{3} J_2$$

and the parameter, $J_m$, is derived from the Lode angle, $\theta$, by:

$$J_m = \sin 3\theta = \frac{3 \sqrt{3} J_3}{2J_2^{3/2}}$$

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In compression tests, $J_m = 1$, whereas in extension tests, $J_m = -1$. This allows to distinguish the yield behaviour in both cases.

Three domains can be considered the $(p', q, J_m)$ space (Fig. 10): (1) a domain, D1, where porosity and permeability decreases moderately due to the closing of pre-existing cracks; (2) a domain, D2, where crack porosity and permeability increase, but where the degree of initial anisotropy does not markedly evolve; and (3) a domain, D3, where hydraulic properties become significantly anisotropic. The first two domains are separated by the plastic yielding curve whereas the last two domains are separated by the failure envelope. These boundaries can be investigated by proper stress path probes.

Acknowledgements

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Application of Slug and Pulse Test Deconvolution  
for Describing the Behaviour of Gas-Water Systems  
A New Approach for Determination of Saturation Conditions

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Abstract

The present work extends the application of slug test deconvolution (PERES et al. 1989, ENACHESCU et al. 1996 and CHAKRABARTY et al. - in print) to gas-water systems. The approach used is to compare injection and withdrawal events by means of Test Type Comparison Curves. It is shown that the method described is sensitive to the gas saturation conditions of the tested formation. Finally, the method is applied to three field examples.

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INTRODUCTION

Hydraulic packer testing is currently the most commonly used method to determine the characteristics of fluid flow in geologic formations. Constant rate tests and slug tests are the most commonly used test methods.

Slug tests are often preferred to constant rate tests due to their relatively short duration and low hardware requirements. However, in many cases slug tests are not the most appropriate method for characterising the formation. The uncertainties often mentioned in connection with slug tests are:

- uncertainties concerning the radius of investigation of a slug test
- unsatisfactory accuracy in the determination of formation transmissivity
- difficulties in the identification of the flow model
- difficulties in determination of the initial formation pressure.

The present work treats slug tests in a different way and demonstrates a new approach for the analysis of these tests. Following concepts are demonstrated:

- Both constant rate and slug tests based on the same theoretical background. Theoretically, there is no difference between the two test methods as far as the derivation of hydraulic parameters is concerned.

- Slug tests yield under certain circumstances more reliable results when analysed using the new deconvolution method.

- The above mentioned disadvantages of slug tests can be overcome.

The deconvolution analysis of slug tests allows the independent determination of storativity and skin factor from a single borehole test. Note that this is not possible from any other test types.

The second part of the work describes the application of the deconvolution technique on numerically simulated test data, for the analysis of gas-water flow systems. Finally, the new approach is applied to three field examples.

THEORETICAL BACKGROUND OF SLUG TEST DECONVOLUTION

Starting from the classic slug test analysis of COOPER/RAMEY, it can be shown that by numerical integration of the dimensionless slug pressure (the RAMEY plot - Fig. 3) an equivalent constant rate response can be calculated (see Figs. 4 and 5). One of the main advantages of a slug test is that the wellbore storage coefficient during the test is well determined. This allows for calculation of the amount of fluid the test interval can store when changing the pressure in the system by a certain value. The wellbore storage effect can be mathematically removed from the test response by using the rate normalisation method (PRES et al. 1989), a process usually called deconvolution. As illustrated in Figures 1 to 7, the formation behaviour can be recognised much earlier in the deconvolved slug than in the constant rate test response. While the constant rate data is still distorted by the wellbore storage effect (the unit slope line in Fig. 7), the deconvolved slug already shows the semi-logarithmic infinite acting straight line approximation, which enables the calculation of formation transmissivity.

The deconvolved slug test proves to be better suitable for the flow model recognition for cases with extensive wellbore storage effects.
Parameter Study

A parameter study should demonstrate the sensitivity of slug test deconvolution method with respect to changes in formation parameters. Figures 8 to 10 present the sensitivity of a simulated slug test to changes in the formation transmissivity, storativity and skin factor at the well. The following conclusions can be drawn:

- Changes in the formation transmissivity influence the vertical position of pressure and derivative data when plotted in log-log coordinates.
- Changes in the formation storativity influence the horizontal position of pressure and derivative data when plotted in log-log coordinates.
- Changes in the skin factor of the wellbore influence the shape of pressure data. The shape and position of pressure derivative remain unchanged.

Following these observations, a type curve set was developed (see Fig. 11). It allows the independent determination of the hydraulic formation parameters (i.e. transmissivity, storativity and skin factor). PERES et al. 1989 show that the principle of superposition is applicable in connection with this method. This enables historical pressure gradients in the formation prior to the test to be accounted for. The method can be applied in connection with more complex flow models as well.

BEHAVIOUR OF GAS-WATER SYSTEMS

The results of characterisation of two-phase flow behaviour of potential nuclear waste repository host rock provide a significant input to performance assessment activities. In the past few years, significant progress has been achieved in both field measurement methodology and theoretical background. However, two-phase flow behaviour, and in particular testing and analysis tools and methods are far from being perfect and thus require further research and development.

The objective of this work is to investigate to what extent a comparison of responses from several different test events can provide additional understanding of two-phase systems. For this purpose, a system has been developed, that allows the user to run numerical simulations (using ECLIPSE 100) for various test sequences under single- and two-phase conditions.

This work concentrates on the initial gas saturation of the formation; this being only one out of a large number of parameters, which are of interest in two-phase flow systems.

The simulations of gas-water systems in this study fulfill the following assumptions:

- flow functions as relative permeabilities and capillary pressures;
- \(pVT\)-phenomena as pressure-dependant gas-water solution and re-solution;
- physical processes both in formation and wellbore are considered.

Simulation Strategy

At this stage the investigations include pulse and slug tests. These tests are commonly used in site characterisation programmes. The reasons for choosing this type of test procedures are provided below:

1. The use of deconvolution provides a powerful tool for diagnosing the behaviour of the test.
2. For single-phase conditions, pulse and slug tests are similar from the theoretical point of view. However, they are different in nature if gas is present. Gas escapes from the system through the test string during a slug test, but is comprised in the system during a pulse test.

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3. Slug and pulse tests are relatively easy to conduct and control for hydrological investigations.

Table 1 summarises the simulation input data for the reference base case.

Five two-phase cases were defined, with a variation of the initial gas saturation of the formation. Table 2 provides the two-phase $pVT$ and flow parameters, which are of relevance to the simulation runs (Fig. 12).

The initial conditions for the five cases are summarised below:

- **Case 1**: water saturated; no free gas initially; $S_g = 0$
- **Case 2**: critical gas saturation; $S_g = 0.01$
- **Case 3**: $S_g = 0.1$
- **Case 4**: $S_g = 0.25$
- **Case 5**: $S_g = 0.5$

For cases 1 to 5 the wellbore fluid was initialised without free or soluted gas.

[Case 0: $S_g = 0$, $p_b = 0$ (single-phase reference case)]

SW, SI, PW, and PI test events (see Nomenclature) were simulated for each of the five cases. The events were simulated with a pressure difference of 2000 kPa. The initial conditions of all events are identical for each case. Also, no pre-test pressure history for the individual test cases was assumed.

**Results**

The results of the simulations were analysed in two steps. First, all simulated test events were deconvolved and plotted together in log-log coordinates for a visual comparison (see Fig. 13, 15 and 17). Second, Test Type Comparison Curves (TTCC) were defined and produced. These curves are calculated by subtracting the deconvolved log-log responses of two different test types, which were conducted under the same initial conditions. These differences are plotted vs. log time. The idea behind these curves is to visualise the differences of the formation response under different inner boundary conditions (test types - see Fig. 14, 16 and 18).

As long as no free gas is present (Case 1), a different system behaviour is seen during injection and production tests. In case of injection, the system acts as single-phase, because no free gas can be dissolved and there is no significant difference in the physical behaviour of dead and saturated water (SI, PI in Fig. 13 and 14). Production tests result in gas coming out of solution which increases the system compressibility. In the early time of SW, the production of a small amount of gas dissolved in the vicinity of the wellbore appears as a positive skin effect, followed by a middle-time stabilisation period, and small gas production in the late time of the test (SW, PW in Fig. 13). The production of gas can be seen as a deviation between SW and PW (Fig. 3).

If small amounts of free gas are initially present (Case 2), the additional gas coming out of solution has no major impact on the middle time behaviour (Fig. 16). Again, the effects of degassing are visible in the early and late time of SW (Fig. 15).

Higher gas saturations lead to different system behaviour (Cases 3 to 5, Case 4 shown in Fig. 17 and 18), with an increase in the difference in responses of injection and production tests (Fig. 18). Due to capillary forces, gas is transported from the formation into the wellbore. The bottom hole pressure no longer converges to the initial pressure but to the threshold pressure (= initial pressure + air entry pressure). During early time of a test, the most important impact on the system is the pressure anomaly caused by initiation of the slug or pulse event. After some time, the capillary forces become dominant, which is visible as a sudden change in the slope of the deconvolved pressure (Fig. 17). During slug tests, large amounts of gas (as compared to Cases 1 and 2) are being produced, and the systems heads to a new stabilisation level. During pulse tests, the gas saturation of the wellbore increases, which leads to a similar, but smoother shape of the curve (SW, PW of Case 4 in Fig. 17).
Figures 19 to 22 present the TTCC-Plot. We compare the different cases for each of the two test types. There is little difference for injection tests, which shows little sensitivity to the initial gas saturation (Fig. 19).

Production tests show big differences between slug and pulse responses, especially for the late time data, which are sensitive to the initial gas saturation (Fig. 20).

Remarkable differences can be recognised between injection and production test data for pulse as well as for slug tests (Fig. 21 and 22). The comparison of these test events gives relatively precise information about the initial gas saturation for the given set of initial conditions.

Note that small amounts of gas (Case 2) can only be detected by comparison of SW and SI, the comparison of other test events provides only limited information (Case 2 in Fig. 21 and 22).

FIELD EXAMPLES

The final part of this study points out the practical aspects of slug and pulse tests in gas-water systems with special emphasis on the theoretical aspects presented above. Three examples taken from the Wellenberg Project in Switzerland are discussed. The three tests were conducted under gas-water conditions.

Each example is visualized in four figures:

1. the pressure response used for analysis, measured as bottom hole pressure (BHP), in Cartesian coordinates,

2. the BHP response of relevant test phases compared with pressure in the test string,

3. the wellbore storage normalized log-log deconvolution plot, and

4. the Test Type Comparison Curve(s) (TTCC).

The Test Type Comparison Curves are calculated by subtracting the deconvolved log pressure response of slug and pulse events of each example. As shown above, this plot visualizes the differences in BHP between different test types.

Under single phase water flow conditions, all slug and pulse events of a test sequence theoretically show the same behaviour. This means that the TTCC is constantly zero. Due to the presence of gas, different behaviour of test events is visible, and the influence of gas on the pressure response of the system leads to deviations of the TTCC from the zero level.

Example 1

The test discussed in Example 1 (Figures 23 to 26) was conducted in a lower Cretaceous marl formation at the Wellenberg site. The SW and PW events were compared. Gas occurrence is evident during the SW phase. This is demonstrated by the sudden pressure change and unstable BHP behaviour. This phenomena is known from the numerical simulation of degassing wellbores and is interpreted as the “breakthrough” of free gas into the formation. The gas is possibly released due to a pressure drop in the formation. Capillary forces, and with gas saturation increasing gas mobility, influence flow behaviour towards the wellbore. Here the gas is produced against atmospheric pressure through the test string.

The “breakthrough” of the free gas is also visible as a sudden change in the slope of the deconvolved pressure. The difference between SW and PW is emphasized in the TTCC.
The pre-test history is important for the test performance. The tests events start at complex pressure and free gas distributions in the reservoir. Therefore, it should be realised that the results of the theoretical appraisal are not directly comparable with the field data yet.

**Example 2**

The second example (Figures 27 to 30), measured at the Wellenberg site as well, compares several slug and pulse events conducted as part of a complex two-phase flow test design (see Fig. 27). Again, pre-test history has a not yet quantified influence. Gas presence is obvious when comparing the BHP and the test tubing pressure during the SW and PW2 phases (Fig. 28).

The change in slope of the SW phase response can be correlated with the gas “breakthrough” into the wellbore. After this point the water flow rate decreases rapidly towards zero resulting in a constant BHP. During the pulse events the gas produced remains in the test interval causing a completely different test response. The TTCC Plot underlines the difference of the slug-event, compared to the pulse tests. While the TTCC-values of the three pulse tests are in the range of zero, the influence of gas, especially in the late time test data, is visible through the deviation of the SW-pulse TTCC from the zero-level.

**Example 3**

This example compares again a SW and a PW (Figures 31 to 34). There is a relatively small difference between the normalised responses of the two phases. This is obvious from the relatively flat TTCC curve. However it is not clear yet as to what extent a flat and near zero TTCC curve is an indicator for certain low gas saturations. No clear interpretation of this case can be done with the new method at the moment since more theoretical work is needed.

**CONCLUSIONS**

A new method for slug test analysis was presented. The advantages as compared with the COOPER/RAMEY method are listed below:

- saving of test time
- improved model recognition
- improved determination of transmissivity
- calculation of storativity and skin from a single well test

There are two limitations of the method:

- the first limitation is related to the ability of the method to account for historical effects. The historical effects proved to be of crucial importance in low permeability formations. Although the application of the method in connection with the principle of superposition was proved to be possible, it has not yet been implemented.

- the radius of investigation of a test depends on the formation transmissivity, storativity and on the test duration. In the case of a slug test the maximum test duration is practically limited by the resolution of the pressure gauge. For certain combinations of permeability range and wellbore storage constant, this leads to a disadvantage of slug tests as compared to constant rate tests.

The pressure response of a homogenous gas-water reservoir with different initial gas saturations has been investigated under different inner boundary conditions (SI, SW, PI, PW). The cases were simulated using the numerical “black oil simulator” ECLIPSE 100. The single-phase problem was used as a reference case. Only the influence of initial gas saturation was investigated.

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The simulation results were deconvolved and plotted as a TTCC-Plot. This type of analysis proved to be sensitive both with respect to the different initial conditions and to the different test types. This new methodology, applied in conjunction with an appropriate test strategy, should allow for quantitative determination of initial saturation conditions in the test zone. More work is necessary to:

- enhance the accuracy of the method,
- extend the application to other test types and, consequently
- further develop and optimise testing strategies for the two-phase gas-water systems.

The formerly developed slug deconvolution method and the TTCC-plot is applied on three field examples, where the occurrence of gas is presumed.

The methods correlate qualitatively with the results of numerical simulations. Especially for gas dominating the pressure response in the wellbore at a certain point of time, the changes in these two plots are clearly visible as changes in slope of the deconvolved pressure and significant deviations from the zero-level of the TTCC-plot.

A method to parameterize these differences is not yet available, but qualitative analysis encourages to former theoretical appraisal and parallel check-up with field data.

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REFERENCES


Fig. 1: Slug Test

Fig. 2: Constant Rate Withdrawal Test

Fig. 3: COOPER/RAMEY Plot

Fig. 4: Integrated Slug Test

Fig. 5: Constant Rate Test; GRINGARTEN/BOURDET Plot

Fig. 6: Deconvolved Slug Test

Fig. 7: Comparison of the Methods

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**Fig. 8:** Influence of Transmissivity

**Fig. 9:** Influence of Storativity

**Fig. 10:** Influence of Skin

**Fig. 11:** Type Curve Set

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<th>Two-Phase Parameters</th>
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<tr>
<td>T 1.00E-09 m2/s</td>
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<td>Porosity 0.01</td>
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Tab. 1: Reference case simulation parameters  
Tab. 2: Two phase flow simulation parameters

**Fig. 12:** Relative permeabilities and capillary pressure [BROOHS & CORREY]
Fig. 19: Test type comparison curves SI-PI

Fig. 20: Test type comparison curves SW-PW

Fig. 21: Test type comparison curves PI-PW

Fig. 22: Test type comparison curves SI-SW

Fig. 23: Example 1 - Pressure response used for analysis

Fig. 24: Example 1 - Downhole pressure response compared with pressure in the test string during the SW phase
Fig. 25: Example 1 - Wellbore storage normalised log-log deconvolution plot

Fig. 26: Example 1 - Test type comparison curve

Fig. 27: Example 2 - Pressure response used for analysis

Fig. 28: Example 2 - Downhole pressure responses compared with pressure in the test string during the SW and PW phases

Fig. 29: Example 2 - Wellbore storage normalised log-log deconvolution plot

Fig. 30: Example 2 - Test type comparison curve
Fig. 31: Example 3 - Pressure response used for analysis

Fig. 32: Example 3 - Downhole pressure response compared with pressure in the test string during the SW phase

Fig. 33: Example 3 - Wellbore storage normalised log-log deconvolution plot

Fig. 34: Example 4 - Test type comparison curve

NOMENCLATURE

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<th>Symbol</th>
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<tr>
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<tr>
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Permeability Anisotropy in Clay-Rich Shear Zones

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Abstract

In actively deforming sediments, fault zones are often observed to form both barriers to and conduits for fluid flow. For example, in the Barbados accretionary complex, long distance lateral fluid migration without vertical leakage is observed along the basal décollement. Thermogenic methane, high fluid pressures and water contents similar in magnitude to their initial depositional state in the underthrust sediments, coupled with a lack of thermogenic methane in the overlying wedge sediments, indicates that the décollement also acts as an aquitard to fluid flow across it. Experimental shearing of clays can go some way to explaining this apparently paradoxical situation.

We report a series of permeability measurements on a consolidated and sheared smectite-rich clay from the Barbados accretionary complex using custom-designed oedometric and ring shear permeameters. The clay, when sheared under fully drained conditions, developed permeability anisotropy with progressive consolidation and shear displacement, increasing from around 15 at 100 kPa effective stress to over 35 at effective stresses of upto 20 MPa. The consolidated clay had a similar level of absolute permeability to that of the sheared clay in the direction parallel to the shear zone. No significant dilation or enhancement of permeability of sheared clay with respect to consolidated clay was observed as the clay was re-sheared on the unloading path. These results suggest that high levels of anisotropic permeability in shear zones cannot be caused by clay fabric alignment alone. Further, since the clay does not dilate strongly during drained shearing, even on a trajectory of decreasing effective stress, some other mechanism is required to explain enhanced fluid flow observed along the décollement of the Barbados accretionary complex.

Distinct fabrics and consistent are produced by shearing of clays to the residual state. The development of Riedel shear arrays is noted in the Barbados clay, together with later throughgoing Y-shears, parallel to the overall shear zone. These shear fabrics play a significant role in the development of high permeability anisotropy in this clay.
Introduction

The significance of the interaction between tectonic and hydrogeological processes in active continental and marine environments has long been appreciated, dating from the ideas of Hubbert and Rubey [1], who demonstrated the role of high fluid pressures in the movement of thrust systems. Since this time, field and laboratory studies have suggested that shear zones in clay-rich sediments can both enhance and retard fluid flow. Faults in un lithified, clay-rich sediments are generally regarded as aquitards, and experimental studies have demonstrated that tectonised clays have low intergranular permeabilities [2, 3, 4, 5, 6, 7]. Clay-rich fault zones in the aseismic portions of accretionary wedges, however, commonly act as conduits for migrating exotic fluids [8, 9]. At the wedge toe of the Barbados accretionary complex, thermogenic methane in the migrating pore fluids indicates a source beneath the wedge 40-60 km distant from where the anomalies were observed [10]. Localised dilation [11], anomalies of temperature [12] and pore water geochemistry [10] in fault zones indicate episodic variations in fluid flow and fluid pressure. Lateral migration of exotic fluids along the décollement appears to be in direct conflict with the theory that clay-rich fault zones act as aquitards. However, hydrogeological modelling suggests that permeability around the décollement of the Barbados wedge must be highly anisotropic, with fault-parallel permeabilities being 3-5 orders of magnitude greater than that of the surrounding wall sediments to permit the lateral flow of fluid that has been observed [9]. Clearly, there must be mechanisms that transform tectonized muds from aquitards into regions of focused flow. We report new results from two fundamentally different types of experiments which show that shearing muds does not cause them to become conductive to fluids, even when such deformation occurs under greatly reduced effective stresses. It seems that only the development of hydrofracture systems can allow muddy faults to be conductive to fluids.

Experimental Techniques

As both mineralogical composition and stress environment both strongly influence permeability evolution through their effects on clay fabric, we report here on a parallel series of consolidation and shear experiments upon a smectitic clay in oedometric permeameters and ring-shear permeameters. These tests have allowed a direct comparison to be made between the stress-strain-permeability behaviour of a clay during one dimensional consolidation and shearing of the same clay to its residual state. Thus we are effectively comparing the permeability of sediments in a shear zone (ring shear) with the behavior of sediments adjacent to a shear zone (oedometer).

Full details of the experimental procedure have been reported elsewhere [5, 6], so only a brief outline will be given here. The oedometer used in this study has been adapted to measure vertical and radial permeability during one dimensional consolidation, using the constant-rate flow pump technique [13], conducted against a constant back pressure of 300 kPa, to ensure saturation of the sample [14]. The ring-shear permeameter [5, 6] has two different cell configurations, with the same dimensions to obtain shear-parallel and shear-normal permeability measurements. Leakage of pore fluid is prevented by double O-ring seals, confining pressurised silicon grease (600 kPa); this also provides lubrication for rotation of the top platen of the ring shear device. The sides of the annular chamber are smooth; the upper and lower sample platens are roughened to induce slip within the body of the sample. A load was applied before shearing commenced, which was then continued at a shear strain rate of $ca. 3.65 \times 10^{-4} \text{ s}^{-1}$ for a time period of up to six days. An insulated cabinet combined with an electronic temperature control system capable of maintaining constant temperature to within 0.5°C enclosed the ring shear experiments. The Barbados clay comprises $> 80\%$ smectite [15].
Results

Fabrics

The fabrics of the sheared clay have distinct elements which are illustrated schematically in figure 1 [16]. Macroscopically, Barbados clay splits along a highly polished and finely striated detachment plane within the body of the sample, parallel to the shear zone walls. SEM photomicrographs (Fig. 2a) of part of the sample above the detachment illustrate multiple sinuous shears cutting through the clay at orientations of between $0^\circ$ and $25^\circ$ to the horizontal. These slip surfaces comprise highly oriented clays and are polished and striated (Fig. 2b and c). A fabric comprising streaked-out clay aggregates can be seen, steeply inclined ($> 60^\circ$) to the shear zone (Fig. 2c). This fabric is divided into individual “packets” by the sinuous shear surfaces noted above, but the fabric within each individual packet retains a constant orientation with respect to the main shear zone.

The main detachment surface (not illustrated) is a Y-fabric element (figure 1), comprising a thin seam of highly aligned clays and represents a slip surface which has resulted in localised deformation during residual-state shearing [17]. The sinuous synthetic low-angle shears are $R_1$ Riedel shears (figure 1) which curve into a Y-orientation parallel to the shear zone margins. The steeply inclined antithetic clay aggregate alignment is in an $R_2$ Riedel-shear orientation (figure 1). $R_1$ shears can only usually accommodate a small degree of slip due to their inclined orientation with respect to the overall displacement of the shear zone [16, 18].

Figure 1. Schematic diagram illustrating the orientation of shear fabrics in the Barbados clay in relation to the strain ellipse (modified from [16]).
Figure 2a. Photomicrograph of sheared Barbados clay, showing numerous sinuous Riedel shears cutting through the clay in an orientation synthetic to the overall sense of shear.

Figure 2b. Photomicrograph illustrating the polished and striated nature of the $R_1$ surfaces. The $R_2$ surfaces are also visible at a high angle to the overall shear zone.
Figure 2c. Photomicrograph clearly illustrating the relationship between the $R_1$ and $R_2$ shears. The contact is sharp with no drag along it, indicative of brittle fracture.

Figure 3. Photomicrograph of oedometrically consolidated Barbados clay. Fabric elements are distinctly different to that of the sheared clay, with regular aggregations of clays visible and a lack of fracturing.
However, the high degree of polishing and continuous striae on R1 surfaces indicate a considerable degree of movement along them. Lack of particle drag associated with these planes indicates that they are discrete, brittle failure surfaces.

The fabric of the oedometrically consolidated Barbados clay is distinctly different (figure 3). The fabric is open in places, with a wide distribution of apparent pore sizes. Domains and aggregates of clay particles are clearly visible, although they do not have a common orientation. In comparison, the sheared sample is far more compact, containing a distinct fabric alignment and is cut by many throughgoing brittle shear surfaces.

**Permeability**

The results in this report are expressed in terms of the coefficient of permeability (hydraulic conductivity). Only vertical permeability measurements were made on consolidated Barbados clay (Fig. 4), and these have a similar magnitude to shear-parallel permeability. Other permeability measurements made on consolidated clays from the Barbados accretionary complex indicate little

![Figure 4. Comparison of permeabilities in consolidated and sheared Barbados clay.](image)

anisotropy at *in situ* effective stresses [19]. Shear-normal permeability is consistently lower than shear-parallel permeability, producing levels of anisotropy of ca. 15 between void ratios of 1.3 and 1.0, rising to 35 as void ratio decreases to 0.7. Figure 5 illustrates the change in void ratio of the clay.
Figure 5. Swelling of consolidated and sheared Barbados clay under decreasing effective stress conditions. The sheared clay shows no enhanced dilation over that of the consolidated clay during reshearing as effective stress decreases.

Figure 6. The hydraulic conductivity and its anisotropy of the sheared Barbados clay remains approximately constant as effective stress decreases, while the hydraulic conductivity of the consolidated clay increases slightly.
as it was resheared under decreasing effective stress conditions. It is evident that the sheared clay has a similar unloading trajectory to that of the consolidated clay. Figure 6 illustrates the effect of reducing effective stress on the hydraulic conductivity of the consolidated and sheared Barbados clay. No enhancement of the hydraulic conductivity is evident along the shear zone, and the level of permeability anisotropy remains approximately constant. The hydraulic conductivity of the consolidated clay increases slightly at low effective stresses.

**Discussion**

The behaviour of high-strain shear zones is complex and is governed by the residual properties of sheared sediments, which, in turn, are dependent on clay content and composition [17]. Three types of shear behaviour have been identified. *Turbulent shear* characterises sediments with clay contents < 20% and is dominated by rotation and rolling of rotund grains, generally resulting in a lack of preferred grain orientation. A *transitional shear* mode exists from 20-50% clay content, where parts of a shear zone experience turbulence as others undergo sliding shear. This may result in formation of clay grain alignments, which subsequently are broken up by turbulence. *Sliding shear* will dominate behaviour of sediment with > 50% clay content and is associated with well-formed polished slip surfaces comprising strongly oriented clays [17], seen clearly developed here as R₁ Riedel shears in the Barbados clay. Similarities are evident on comparing the fabrics developed in the Barbados clay in the ring shear permeameter with those in natural and artificial quartz and calcite fault gouges described previously [16, 18]. The strength and the differences between the structural elements are somewhat different in the Barbados clay and the quartz gouges, probably as a result of greater cohesion and lower interparticle friction in the smectite-rich Barbados clay. Overall similarities are compelling, which leads us to an interpretation of fabric evolution that is similar to that suggested previously [16]. P-fabrics are associated with shear-oblque compression (figure 1); R₁ fabrics are interpreted as relatively early formed features, accommodating small increments of slip and the main Y-element at the base of the deformed zone as the major slip plane which breaks through after a few tens of degrees of platen rotation. This boundary is marked by a thin seam of compact and highly aligned clays, which represent a slip surface which localised the main displacement during residual state shearing [17].

**Effects of Shear Fabrics on Permeability Anisotropy**

The results of this and previous experimental programmes indicate that permeability anisotropy generated in sheared clay probably rises with increasing effective stress and shear strain. Clay content and grain size of the clays also affect the magnitude of permeability anisotropy, which is probably a direct reflection of the intensity of the shear fabric formed during deformation. This study and others [20, 21, 22] suggest that deformation of clay-rich sediments tends to result in formation of discrete shear zones, which generally become more closely spaced as particle size decreases and clay content increases. The Barbados clay attains a higher permeability anisotropy, has a higher density of shear surfaces and a finer mean grain size than kaolinite and silty clay for which similar experiments have been conducted [6, 7].

Comparison of shear zone permeability with that of the corresponding consolidated sediment indicates that although the shear-parallel permeability is greater than the shear-normal permeability, it is of a similar magnitude to that of the consolidated sediment. Together with fabric analysis, this suggests that permeability anisotropy of sheared clay is caused by reduction of shear-normal permeability in these experiments and that intergranular permeability is not enhanced parallel to the shear zone (we believe that the fractures observed in these clays were
closed during the static permeability tests under vertical load, and that we measured intergranular permeability). Strain localisation may have resulted in enhanced pore collapse in this part of the fabric, resulting in lower permeability across the shear zone. Furthermore, the strong alignment of clays to form the R₁ Riedel shear surfaces may form a more effective barrier to fluid flow across the shear zone than along it.

Geological and Geotechnical Implications For Fault-Related Fluid Flow

Our results have implications for a wide range of geological and geotechnical investigations of phenomena as diverse as landsliding, tectonic faulting, aquifer quality, petroleum migration and trap formation. For example, scaly fabrics and anastomosing R₁ shear surfaces are observed in the basal slip surfaces of landslides, which often have abnormal water contents and act to obstruct and focus drainage, affecting the stability of the slide [23]. The décollement of the Barbados accretionary complex is located in a clay-rich section comprising at least 80% smectite [15] and is expected to deform predominantly by a sliding-shear mechanism. Our experiments suggest that this would result in a high degree of permeability anisotropy as a result of reduction of vertical permeability of the shear zone. Low vertical permeability of the décollement can account for the retention of exotic fluids during lateral flow within this structure by inhibiting leakage into the surrounding sediment. Recent packer tests [24] and modelling [25, 26] confirm that the vertical permeability of the décollement of the Barbados accretionary complex is extremely low at ca. 10⁻¹² ms⁻¹. Our results for permeability across the shear zone are quite consistent with in situ and modelled results.

It is interesting to note that fractures developed experimentally in Barbados clay have a similar morphology and orientation to scaly fabrics located in the décollement of the Barbados accretionary complex [27]. Significantly, however, there was no enhancement of permeability along the shear zone under any of the experimental stress conditions investigated. Soil mechanics theory [e.g. 28, 29] and geological research [e.g. 30] suggest that dilation may be important in sheared granular sediments and overconsolidated clays. Increased water contents in sheared overconsolidated clays have been observed where the effective stress is reduced prior to failure [23]. This testing programme included a series of permeability tests on overconsolidated silty clay sheared to the residual state after incremental unloading from the peak vertical effective stress to simulate the effects of a rise in pore pressure or removal of overburden. Weak dilation and strain softening were evident in the shear strength response of the sample, but neither the void ratio nor the permeability increased above that recorded during elastic rebound of the consolidated clay. Furthermore, the levels of permeability anisotropy attained at the highest average vertical effective stresses were generally conserved during unloading. In contrast, the permeability measured along the décollement during the packer tests is far higher than shear-parallel intergranular permeability documented in our tests [24]. It seems therefore, likely that fracture pathways must be open sub-parallel to the décollement as a consequence of elevated fluid pressures to account for the high permeabilities observed in the natural system [e.g.; 5, 11, 31, 32].
Conclusions

Levels of absolute permeability following consolidation of this clay were comparable with others reported from the Barbados accretionary complex [19]; permeability of the sheared clay along the shear zone was similar to that of the consolidated sediment. Permeability across the shear zone was up to 35 times less than along it. The results suggest that effective barriers to fluid migration can be produced by shearing in clays and that permeability anisotropy is probably dependent on shear displacement, clay content and mean grain size. Localisation of strain during residual state shearing produces a local volume loss and permeability decrease across the shear zone in the Barbados clay. The alignment of the R₁ fabric at a low angle to the direction of shear and the strain localisation associated with these slip surfaces explains the levels of absolute hydraulic conductivity and magnitude of permeability anisotropy that was measured in these experiments. It appears that grain alignment fabrics are unlikely to contribute significantly to the overall anisotropy of the sheared silty clay samples. In general, it appears that the production of localised zones of greater compaction and lower permeability is more effective in producing permeability anisotropy than the realignment of clay minerals alone. In natural shear zones, the anastomosing nature of slip surfaces may result in even more extreme permeability anisotropy, which can explain both fluid retention in clay-rich shear zones and their behaviour as effective aquitards. However, it would appear that to enhance the effective permeability along clay-rich shear zones by 3–5 orders of magnitude requires hydrofracturing and consequently, near-lithostatic fluid pressures.

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References


Compaction of TOC-rich Shales Due to Kerogen Conversion: Implications for Fluid Flow and Overpressure

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Abstract:

TOC-rich shales (10 % TOC) have been artificially matured at temperatures between 200 and 350 °C under controlled axial stress (20 - 40 MPa) for up to 350 hours. The volume change of the cylindrical samples was monitored continuously throughout the experiment. The performed experiments showed that the compaction associated with the thermal decomposition of the kerogen is significantly larger under hydrous than under dry pyrolysis conditions. This observation points at an important role of water in the conversion of kerogen.

Semi quantitative permeability tests indicated that sample permeability had decreased at least one order of magnitude after the compaction pyrolysis experiments. This permeability reduction in combination with the observed compaction is the most probable mechanism for overpressure formation in TOC-rich source rock sequences.
Introduction:

Pressure-driven volume flow of a separate hydrocarbon phase is now the most widely accepted mechanism for primary migration and expulsion of petroleum from source rocks. This process influences all subsequent steps resulting, eventually, in the accumulation of petroleum in reservoirs. It is therefore necessary to obtain a comprehensive understanding of its timing and the driving forces. In order to address these problems from an experimental point of view an automatic set-up was constructed which permits the conduction of laboratory experiments under elevated temperature and pressure conditions. One advantage of this set-up is its capability to conduct different migration experiments, one after the other, with the same rock sample.

Experimental:

Figure 1 shows a scheme of the triaxial cell, which was developed in our laboratory for the simulation of subsurface temperature and pressure conditions. In this flow cell a cylindrical rock sample (E), sandwiched between two porous stainless steel plates (F), is placed between two pistons which exert a controlled axial pressure (lithostatic pressure, L). Sample and pistons are coated with a temperature-resistant two-layered sleeve to which a radial confining pressure can be applied (tube D). Additional conduits (C) inside the pistons are used to control the fluid pressures on the upper and lower side of the rock sample independently.

The entire flow cell, can be heated under controlled pressure conditions to temperatures up to 350 °C. Fluid pressures can reach over 300 bars and the lithostatic pressure is limited mainly by the mechanical stability of the rock sample. With this kind of equipment experiments have been performed which are referred to as “compaction experiments”.

Compaction experiment

In the compaction experiment reported here an immature source rock sample (Toarcian shale source rock, Hils Syncline area, NW Germany, Rm=0.53%) was matured artificially for 80 hours at 347 °C under constant axial stress (220 bars, no radial pressure). The length of the sample was monitored throughout the experiment by a dilatometric transducer. Contrary to compaction pyrolysis experiments with powdered source rock performed by Takeda et al. (1989) the experiments use whole rock samples and constant axial load. By carefully controlling the pressure during the heating and cooling phase of the experiment, the matured rock sample could be recovered undamaged after the end of experiment and used for further investigations.
Figure 2. Variation of sample thickness during a compaction experiment with a rich Toarcian shale source rock.

Figure 2 shows the decrease in sample thickness for the rich (10 % TOC) Toarcian shale source rock in this compaction experiment. After an initial phase of thermal expansion of the entire experimental set-up (0-1h) the sample thickness continuously decreases. The effect is reproducible and the samples have been checked after the experiment to make sure, that there is really a change in sample volume (and not only a radial deformation of the sample). Semi quantitative permeability tests indicated that sample permeability had decreased at least one order of magnitude after the compaction pyrolysis experiments.

Similar samples with lower TOC-content (1.5 % TOC) show no compaction. The compaction rate is temperature dependent and there is a linear relationship (Fig. 3) between the observed compaction and the transformation ratio of the kerogen, which had been calculated from the data of hydrous pyrolysis experiments (Marzi 1989).

Figure 3. Comparison of compaction and kerogen transformation ratio (TR) calculated from hydrous pyrolysis results for a Toarcian shale source rock
The amount of compaction depends on the pyrolysis conditions. As shown in Fig. 4, hydrous pyrolysis results in a significantly higher compaction and a larger expulsion of organic matter.

**Figure 4. Comparison between different pyrolysis techniques**

**Discussion**

Based on these experimental results, it can be concluded that kerogen in organic matter-rich source rocks is load-bearing (Figure 5). Therefore, during maturation, compaction occurs not only due to loss of original porosity but also by conversion of load-bearing kerogen into bitumen which is extruded into the (usually water-filled) pore space. Therefore, kerogen conversion causes either a fluid flow in the rock or overpressuring.

With the present knowledge of the kinetics of kerogen conversion computer modelling of this effect seems possible and promising.

**Figure 5. Model of compaction due to kerogen conversion**
Literature:


Modelling Coupled Gas and Water Flow along Preferential Pathways in Low Permeability Media

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Abstract

The Darcy two-phase flow model is widely used in simulating the migration of gas and water in subsurface environments. The model is a generalization of the classical Darcy model for groundwater flow in a porous medium, and is a continuum model based on the concept of there being a representative elementary volume over which water and gas flow properties can be averaged. There is however, some debate about the validity of using continuum Darcy models to simulate gas migration in very low permeability media, such as the argillaceous formations considered for radioactive waste disposal. This is because in such media gas will tend to migrate along a small number of preferential pathways ("paths of least resistance"), which is not consistent with a continuum interpretation. The question thus arises of what alternative models are available.

One alternative is to use a capillary bundle approach. An ensemble of straight capillaries provides a direct topological representation of the gas migration pathways in a medium. The ensemble consists of capillaries with different radii, with the radii distributed according to a probability distribution function. Capillary bundle models exhibit hysteresis behaviour because the gas permeability of the medium at a given saturation level is dependent on whether saturation is increasing or decreasing. This type of behaviour is observed in experiments, but can only be reproduced in continuum Darcy models if they are explicitly formulated with history-dependent parameters.

A natural extension of the standard capillary bundle model is to consider geomechanical effects. In the standard model each capillary pathways is of constant radius, but in the geomechanical model the radii can be time- and space-dependent functions of gas pressure and/or water pressure. The underlying simplicity of the capillary bundle approach is such that it is relatively straightforward to incorporate
geomechanical relationships between the pathway radii. This paper presents the results of extending the capillary bundle approach to incorporate generic gas migration behaviour, in which pathways are dynamically opened as the gas pressure exceeds a local threshold value, propagate through the material and close if the gas pressure drops. This three-stage behaviour:

1. threshold
2. propagation
3. relaxation

is typically observed in gas breakthrough experiments in argillaceous media. It produces episodic gas outflux histories for constant gas influx and hysteresis phenomena.

Further extensions of this approach to modelling gas migration in preferential pathways are also discussed. The first objective is to work in close collaboration with experimentalists to determine the material-specific values for the parameters in the generic model presented here.

1. Introduction

There is currently much debate about the way in gas migrates through low permeability media, such as the argillaceous formations considered for radioactive waste disposal [1]. Experimental studies indicate that in such media gas will tend to migrate along a small number of preferential pathways ("paths of least resistance") and will exhibit episodic and hysteretic behaviour [2]. Gaining a quantitative insight into this behaviour, via mathematical modelling, poses significant challenges. The traditional Darcy two-phase flow model is widely used in simulating the migration of gas and water in subsurface environments. The model is a generalization of the classical Darcy model for groundwater flow in a porous medium, and is a continuum model based on the concept of there being a representative elementary volume over which water and gas flow properties can be averaged. The use of a continuum model to gain an insight into spatially discrete is very much open to question, so attention has recently been focused on what alternative models are available.

One alternative is to use a capillary bundle approach [3]. An ensemble of straight capillaries provides a direct topological representation of the gas migration pathways in a medium. The ensemble consists of capillaries with different radii, with the radii distributed according to a probability distribution function. Capillary bundle models exhibit hysteresis behaviour because the gas permeability of the medium at a given saturation level is dependent on whether the water saturation is increasing or decreasing [3]. The standard capillary bundle model does not however exhibit episodic behaviour for constant gas influx. This has been addressed in two ways. First by extending the one-dimensional bundle to a two-dimensional network [4] and secondly by adding a simple capillary closure mechanism which is a function of gas-pressure [5][6]. In both cases, the drop in gas pressure due to gas breakthrough
leads to water re-invading the pathways along which gas breakthrough occurred. This blocks the gas breakthrough pathway, leading to a rise in gas pressure which subsequently again displaces the water from gas breakthrough pathways, restarting the cyclic behaviour. These extended models thus exhibit the characteristic three-stage behaviour:

1. threshold
2. propagation
3. relaxation

typically observed in gas breakthrough experiments in argillaceous media. Each of these effects is however based on the concept of gas having to overcome capillary forces to displace porewater along pre-existing pathways.

In some argillaceous media it could be argued that it is in fact geomechanical effects that are dominant because they control the form of the pathways. For example, the gas may have to overcome the local effective stress to open or widen a micro-fracture. This opening will then be dynamically propagated through the medium, to form a preferential pathway held open by the gas pressure against the local effective stress along the pathway. Finally, when gas breakthrough occurs the gas pressure will drop and the pathway will close under the action of the effective stress. It may also be the case that in some media the gas migration is controlled by a mixture of capillary and geomechanical forces. This provides the motivation for considering a generic extension of the capillary bundle model, which we shall call the Gas Pathway Generation (GPG) model. This paper describes the mathematical formulation of the generic GPG model (Section 2) of a single pathway and discusses some initial results on simple material-specific formulations (Section 3). The paper also briefly considers further extensions to the model (Section 4).

2. Generic Formulation of the Gas Pathway Generation Model

Consider a partly formed gas pathway in a transport medium. For simplicity suppose that the gas moves in the positive \( x \) direction and that the gas pathway is radially symmetric. The GPG model is formulated by considering the forces acting at the interface between the pathway (assumed to be filled with gas) and the transport medium, which includes granular metrival (for example, clay particles) and free or bonded porewater. The position of the gas-medium interface is given by \( y = s(x, t) \) (Figure 1).

Newton’s Second law applied to a small section of the gas-medium interface of length \( \Delta l \) and position \( r \), where

\[
\begin{align*}
\mathbf{r} &= x \hat{x} + s \hat{y} \\
\Delta l^2 &= \Delta s^2 + \Delta x^2
\end{align*}
\]

(with \( \hat{x} \) and \( \hat{y} \) being unit vectors in the \( x \) and \( y \) directions, respectively) yields the following generic equation for \( r \)

\[
m \frac{\partial^2 \mathbf{r}}{\partial t^2} = -\beta \frac{\partial \mathbf{r}}{\partial t} + \mathbf{F}(\mathbf{r}, t)
\]
where $m$, $\beta$ and $\mathbf{F}$ are the mass, drag coefficient and force per unit surface area of the gas-medium interface. The force term indicates the forces that need to be overcome if the gas-medium interface is to move, whilst the drag term controls the rate at which the gas-medium interface propagation occurs.

![Figure 1. Idealised radially symmetric gas-medium interface.](image)

The force per unit area of the gas-medium interface, $\mathbf{F}(r, t)$, can be written as

$$
\mathbf{F}(r, t) = \left(P_{\text{gas}}(x, t) - \sigma(r, t)\right)\mathbf{n}_N + \lim_{\Delta x, \Delta t \to 0} \frac{F_t|_{x=\Delta x/2} - F_t|_{x=-\Delta x/2}}{\Delta t} (2.4)
$$

where and $n_T(x, t)$, $n_N(x, t)$ are the tangential and normal unit vectors to the gas-medium interface:

$$
n_T(x, t) = \frac{x + \frac{\partial s}{\partial x}r}{\sqrt{1 + \left(\frac{\partial s}{\partial x}\right)^2}} (2.5)$$

$$
n_N(x, t) = \frac{-\frac{\partial s}{\partial x}x + r}{\sqrt{1 + \left(\frac{\partial s}{\partial x}\right)^2}} (2.6)
$$

The term $\sigma$ is a generalized threshold tensor, quantifying the forces acting normal to the gas-medium interface that need to be overcome if the gas-medium interface is to move. This will be material-specific and may include

1. the threshold force for gas to displace free porewater (ie. capillary pressure)
2. the threshold force for gas to deform the medium (ie. effective stress)

The third term includes the magnitude of the surface tension per unit length of interface, $F_t$, and acts tangentially to the surface. Thus:

$$
\mathbf{F}(r, t) = P_{\text{gas}}(x, t)\mathbf{n}_N - \sigma(r, t)\mathbf{n}_N + \frac{d(F_t n_T)}{dx} \sqrt{1 + \left(\frac{\partial s}{\partial x}\right)^2} (2.7)
$$

This term may be significant in material such as clay, where a preferential pathway is formed by displacement of clay platelets in a direction parallel to the platelet alignment: moving one end of an individual platelet will cause the other end to also move.
Supposing simple diagonal forms for the drag and the generalized threshold tensor, that is

$$\beta(r, t) = \begin{pmatrix} \beta^\nu(r, t) & 0 \\ 0 & \beta^\sigma(r, t) \end{pmatrix}$$  \hspace{1cm} (2.8)

$$\sigma(r, t) = \begin{pmatrix} \sigma^\sigma(r, t) & 0 \\ 0 & \sigma^\nu(r, t) \end{pmatrix}$$  \hspace{1cm} (2.9)

and assuming constant surface tension, the equations of motion for the infinitesimal section at position r are

$$m \frac{\partial^2 s}{\partial t^2} + \beta^\nu \frac{\partial s}{\partial t} = \frac{\partial F_i \frac{\partial s}{\partial x}}{1 + \left( \frac{\partial s}{\partial x} \right)^2} + \frac{F_i \frac{\partial^2 s}{\partial x^2}}{1 + \left( \frac{\partial s}{\partial x} \right)^2} \frac{\partial P_{gas}(x, t) - \sigma^\nu(r, t)}{1 + \left( \frac{\partial s}{\partial x} \right)^2}$$  \hspace{1cm} (2.10)

$$m \frac{\partial^2 x}{\partial t^2} + \beta^\sigma \frac{\partial x}{\partial t} = \frac{\partial F_i \frac{\partial s}{\partial x}}{1 + \left( \frac{\partial s}{\partial x} \right)^2} - \frac{F_i \frac{\partial s \frac{\partial^2 s}{\partial x^2}} \partial x}{1 + \left( \frac{\partial s}{\partial x} \right)^2} - \frac{\partial s \left( P_{gas}(x, t) - \sigma^\sigma(r, t) \right)}{1 + \left( \frac{\partial s}{\partial x} \right)^2}$$  \hspace{1cm} (2.11)

These equations are coupled with equations of motion for the flow of gas in the pathway itself, which are derived from mass conservation, gas flow and a gas equation of state. Here, the gas velocity, \( v \), is assumed to be governed by Stokes law for slow flow [7] and the ideal gas law is used as an equation of state.

$$\frac{\partial}{\partial t} (\rho s^2) = -\frac{\partial}{\partial x} (\rho s^2 v)$$  \hspace{1cm} (2.12)

$$v = -\frac{\rho_{gas} g s^2}{8 \mu_{gas}} \frac{\partial P_{gas}}{\partial x}$$  \hspace{1cm} (2.13)

$$P_{gas} = \Gamma \rho_{gas}$$  \hspace{1cm} (2.14)

where \( \mu_{gas} \) and \( \rho_{gas} \) are the gas viscosity and density. The equations (2.10)–(2.14) represent the generic formulation of the GPG model. To formulate material-specific versions the \( \beta \), \( \sigma \) and \( F_i \) terms need to be specified by considering the mass and momentum conservation equations for the medium.

3. Material-Specific Examples of the GPG Model

The above formulation is very generic. In order to make progress in modelling gas migration in a given medium, the terms \( F \) and \( \beta \) need to be expressed in more material-specific terms. The examples below show two simple examples of this. It should be noted that these examples are intended simply as demonstrations of the process by which the generic form of the GPG is customized to different media and to indicate how the GPG model provides an excellent framework for incorporating a range of gas migration effects.

3.1 Capillary Pressure Dominated Pathways

In materials such as sand or sand-bentonite mixtures it is likely that the action of capillary forces displacing porewater along pre-existing pathways will dominate the propagation of the gas-medium.
interface. In formulating the material-specific GPG model in this case, advantage can be taken of the fact that the pathways are pre-existing. The term $s$ thus represents the pathway radius, which is constant time but may vary along its length (i.e. $s = s(x)$). In addition, the terms $\beta^x$ and $\beta^y$ in the drag tensor $\beta$ in (2.8) can be approximated by:

$$
\beta^x(r, t) = 8\mu_{\text{water}} \int_X^L \frac{1}{s(x)^2} dx
$$

(3.15)

$$
\beta^y(r, t) = 0
$$

(3.16)

where $X$ is the position of the gas-medium interface and $L$ is the total length of the sample. Similarly, the terms $\sigma^x$ and $\sigma^y$ in the generalized threshold tensor $\sigma$ in (2.9) can be approximated by:

$$
\sigma^x(r, t) = P_{\text{water}} + P_{\text{capillary}}
$$

(3.17)

$$
\sigma^y(r, t) = 0
$$

(3.18)

The position, $X(t)$, of the furthest point of the gas-medium interface is found by solving equations (2.10)–(2.14) with $\frac{dX}{dt} \to -\infty$. Assuming negligible mass and surface tension, a simplified set of equations is obtained:

$$
\frac{dX}{dt} = \frac{P_{\text{gas}}(x, t) - \sigma^x(r, t)}{\beta^x}
$$

(3.19)

which is, in fact, the same governing equation as for a single capillary in the capillary bundle model [3].

3.2 Effective Stress Dominated Pathways

In an argillaceous medium, such as Boom clay, it is apparent that the migration of gas is controlled by the effective stress. A simple approximation to this is to assume that the terms $\beta^x$ and $\beta^y$ are constant and that the terms $\sigma^x$ and $\sigma^y$ in the generalized threshold tensor $\sigma$ in (2.9) can be approximated by:

$$
\sigma^x(r, t) = \Sigma^x
$$

(3.20)

$$
\sigma^y(r, t) = \Sigma^y + \lambda s
$$

(3.21)

where $\Sigma^x$ and $\Sigma^y$ are the components of the effective stress tensor in the $x$ and $y$ directions, and $\lambda$ is a coefficient of elasticity.

Imposing the boundary condition $\frac{dX}{dt} = 0$ at $x = 0$, the equations of motion (2.10)–(2.14) become, for negligible mass and surface tension

$$
\frac{\partial s}{\partial t} = \frac{P_{\text{gas}}(x, t) - \sigma^y(r, t)}{\beta^y}
$$

(3.22)

$$
\frac{\partial x}{\partial t} = 0
$$

(3.23)

These equations lead to the propagation of a gas pathway whose maximum width is given by

$$
\alpha_{\text{max}} = \frac{P_{\text{gas}}(x, t) - \Sigma^y}{\lambda(x)}
$$

(3.24)
For a given gas pressure the tip of the pathway will also move at a constant velocity. Preliminary numerical results showing the propagation of the gas-medium interface under constant pressure conditions are shown in Figure 2. Further development of the numerical solver is currently under way to simulate the gas-medium interface propagation under conditions of varying gas pressure, particularly the case where the gas pressure drops due to gas breakthrough which should cause the interface to move backwards until the pathway is completely re-sealed.

![Figure 2. The propagation of a gas-medium interface overcoming effective stress, showing the interface at constant time intervals.](image)

It is relatively simple to extend the GPG formulation to consider the displacement of free porewater from the medium. If the furthest point of the gas-medium interface \( X(t) \) then the volume of the pathway is

\[
\pi \int_0^{X(t)} s^2(x, t) \, dx
\]  

(3.25)

If the solid particles in the medium are assumed to not compressed by water the volume of water displaced will equal the volume of the gas pathway, so the water flux will be:

\[
\pi \frac{dX}{dt} s^2 + \pi \int_0^{X(t)} \frac{\partial s}{\partial t} \, dx
\]  

(3.26)

Thus the fact that the gas-medium interface propagates at a constant speed means that water will be displaced at a constant rate before gas breakthrough, which is consistent with experimental observations [2]. Further studies focusing on gaining greater understanding of the effective stress and drag terms, and validation of the model by reference to experimental measurements of gas breakthrough times, expelled water volume before gas breakthrough and water saturation at breakthrough are however needed if further progress is to be made.
4. Conclusions and Further Work

The GPG model is based on a very generic formulation of the generation of a single gas pathway and is still in its infancy with regard to incorporating material-specific phenomena. The examples above do however show that it provides an excellent framework for the incorporation of various geomechanical and capillary forces and, as such, should aid the discussion between experimentalists, geologists and modellers that is required if gains are to be made in the quantitative understanding of gas migration through low permeability media.

Theoretically, it should be simple to extend the single pathway model to a multiple pathway model (analogous to the capillary bundle model) but before doing that it is perhaps more important to identify additional material-specific formulations for single pathways. A prototype numerical solver has been developed to solve the generic GPG model equations with a very wide range of threshold and drag terms, so it should be relatively straightforward to undertake simulations of pathway generation in a range of media. Given this facility, the focus then shifts to analysing experimental results in order to determine particular model parameters and examine the validity of the model.

References


Origin of Preferential Clay Particle Orientation in Faults, and Relationships With Pore–Water Flow and Water–Sediment Interactions: Two Natural Examples

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Extended Abstract

We present two natural examples of shear deformation associated with thrust faulting in clayey sediments. The first example is the basal décollement fault of the Barbados accretionary prism (Lesser Antilles), drilled during ODP Leg 156 (June-July 1994). This décollement is an active fault where the relationships between pore–water and deformation can be studied in situ. The second example is the Eocene south–Pyrenean basin (northern Spain), studied by the European Community EBRO Network Working Group (1994-95). In this case, fluid activity in fossil thrust–faults was studied indirectly through the products of water–sediment interactions.

The active Barbados accretionary prism décollement fault

The Barbados accretionary prism is constituted by sediments offscraped from the Atlantic plate at the leading edge of the overriding Caribbean plate. The décollement fault separates the Miocene to Recent sediments involved in thrust deformation in the prism, from the underthrust Late Cretaceous to Oligocene sediments. The prism is mainly made of smectitic mudstone, whereas the underthrust sediments comprise turbiditic silt and clay alternances. During ODP Leg 156, the décollement fault was drilled at 2 and 4 km arcward from the deformation front, at about 500 m below sea floor. In this area, temperature is about 40°C at the level of the décollement.

Fluid overpressuring and undercompaction of the sediments in the décollement fault zone is revealed by modelling of seismic reflection, density logging and packer–test data, and fluid circulation at the top of the fault zone is indicated by chemical anomaly of pore–water (chloride depletion). At the core–scale, the décollement fault zone corresponds to a 40–m-thick interval which exhibits cm–thick, sharply–bounded deformation bands where shear strain and tectonic displacement are concentrated. These bands affect only the clay–rich lithologies. In these bands, deformation is characterised by a “scaly foliation”, i.e. patterns of anastomosing surfaces which divide the sediment into sub–mm–scale lenses. Under the scanning (SEM) and transmission (TEM) electron microscopes, the undeformed clay shows a high (>55%) porosity honey–comb fabric characterised by the irregular distribution of clay particle long–axes, with sub–equant, up to 5 μm polygonal pores. The domains of scaly foliation show strong preferential orientation of clay particle long–axes. The preferentially oriented particles are tightly packed, with pore–size strongly reduced in relation to the undeformed clay, and showing a high length/width ratio (1≤5 μm; w≤1 μm). Fluid circulation along the deformation zones is attested by the occurrence of mm–thick carbonate (rhodochrosite) veins which precipitated along the scaly foliation.
Eocene south-Pyrenean thrust faults

The Ainsa basin was formed during the lower Eocene in the inner part of the South–Pyrenean foreland basin. The basin was filled up with turbiditic marls and siltstones comprising a detritic clay fraction of micas (and/or illite) and chlorite. The sediments were affected by thrusting soon after deposition, i.e. in a poorly lithified state. In the studied area, deformation did not imply formation of a regional cleavage.

Each thrust fault consists of a ten of metres–thick shear zone displaying striated shear surface networks, moderate to intense sigmoidal pressure–solution cleavage and small to medium–scale folds. In the central and most intensely deformed part of the shear zone occurs a few–cm–thick, sharply–bounded shear band with a characteristic shear facies. This facies consists of a black sediment distinct from the surrounding grey marls, and affected by an intense sigmoidal pressure–solution cleavage with numerous calcite veinlets aligned parallel to the cleavage. The veinlets are of two types. The first type is made of cleavage–parallel calcite fibres having intense stylolitisation along their borders; they result from cleavage–parallel stretching of the siltstone layers during shear deformation. The second type of veinlets are late extensional veins not affected by stylolitisation. Two types of water–sediment interactions were involved in shear fabric formation. First, SEM images show that the black sediment is mainly made of preferentially oriented and tightly packed clay particles, and is strongly depleted in calcite grains with respect to the original marl, indicating pressure–solution in the sheared sediment and calcite transfer toward the syn–kinematic fibrous veinlets. Second, XRD and TEM data indicate that the clay fraction in the black sediment consists mainly of dickite, a kaolin polynotype, whereas dickite is absent in the surrounding grey marls comprising only mica (and/or illite) and chlorite. TEM images suggest a mica–dickite transformation by a pressure–solution mechanism. The reaction implies release of potassium, which may have left the system through fluid circulation in the shear zone. This interpretation is consistent with mass transfer calculations (Gresen method) which indicate a depletion of the potassium in the black sediment in relation to the surrounding lithologies.

Discussion

The examples above show that fault–related shear deformation in clayey sediments is characterised by preferential orientation of clay particle long–axes, and this shear fabric may result from different deformation mechanisms. In the case of the Barbados accretionary prism, the clay shear fabric results only from mechanical rotation/distortion of clay particles, whereas in the case of the southern Pyrenees, the clay shear fabric formation also involved water–sediment interactions which affected both the clay fraction (mica–dickite transformation) and other minerals (pressure–solution of calcite grains). These different behaviours were probably controlled by differences of initial textures (>55% porosity in Barbados, probably less in the Pyrenees due to the presence of a silt fraction) and lithologies (>80% of smectitic clay and absence of calcite in Barbados, important silt fraction and about 30% of calcite in the southern Pyrenees). Presently available data make it difficult to assess the possible influence of other factors such as the duration of loading or temperature.

In both cases, the clay shear fabric formation is associated with strong porosity collapse. However, different features (carbonate veins in carbonate–free host–sediment in Barbados, release of potassium during mica–dickite transformation, and second generation of calcite veinlets not associated with local dissolution in the southern Pyrenees) indicate open systems with preferential fluid flow circulation through the fault zones during or after deformation. This indicates that porosity collapse is not necessarily associated with a permanent reduction of permeability, probably due to the fact that preferred clay particle orientation creates potential pathways for drainage of fluids along the
deformation zones. Formation of the rhodochrosite veins in Barbados, and the second generation calcite veinlets in the southern Pyrenees correspond to dilatancy in a direction opposite to the direction of compression responsible for the formation of the foliation. This kinematic incompatibility requires that shear stress was relaxed when the veins formed, probably due to injection of high pore pressure fluid which opened the pre-existing fabric, and decoupled the stress at the boundaries of the over pressured zone.

We thus infer that cyclic variations of stress state controlled by variations in pore pressure occur in the shear bands. Shear deformation is achieved during episodes of relatively low pore pressure and significant shear stress, and results in low permeability of deformation bands, whereas episodes of high pore pressure result in dilation, increased permeability and inhibition of shear strain. Tectonic displacement along the sharp boundaries of the shear bands is likely to be favoured during the high pore pressure episodes, and the shear deformation would account for only part of the cumulative displacement.

These studies point out the importance of the variations of fault permeability through time, but the shape and periodicity of these variations remain largely unknown. Quantification of these variations is an important goal for assessment of fault transport capacities, and needs better knowledge of controlling factors, principally the kinetics of fault sealing mechanisms. This may be realised through quantitative studies of water-sediment interactions and mass transfert balances.
The Brush Model – a New Approach to Numerical Modeling of Matrix Diffusion in Fractured Claystone

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Abstract

A special approach for numerical modeling of contaminant transport in fractured claystone is presented. The rock matrix and the fractures are simulated with individual formulations for FE grids and transport, coupled into a single model. The capacity of the rock matrix to take up contaminants is taken into consideration with a discrete simulation of matrix diffusion. Thus, the natural process of retardation due to matrix diffusion can be better simulated than by a standard introduction of an empirical parameter into the transport equation.

Transport in groundwater in fractured claystone can be simulated using a model called a "brush model". The "brush handle" is discretized by 2-D finite elements. Advection-dispersive transport in groundwater in the fractures is assumed. The contaminant diffuses into 1D finite elements perpendicular to the fractures, i.e., the "bristles of the brush".

The new approach yields the same results for contaminant diffusion in the rock matrix as the standard combination of 2-D and 3-D finite elements. The advantages of the brush model are saving of computer time, especially in large-scale modeling, and a simpler mesh generation. Moreover, it is easier to keep to the stability criteria for the two different transport mechanisms.

The approach is calibrated with results from forced-gradient multi-tracer tests in fractured claystone. In addition, results of laboratory diffusion experiments were taken into consideration. The model was validated with an independent data set. The resulting model matched the history of the current situation at the toxic waste disposal site Münchhagen (Lower Saxony), which is situated in a highly fractured Lower Cretaceous claystone.

Different scenarios with several parameter variations are presented. The conclusion is drawn that matrix diffusion is an important property of fractured claystone for contaminant retardation. This, as well as the hydraulic properties of the geological barrier, has to be taken into account when a permanent repository for radioactive wastes is assessed.
1 Introduction

The assessment of the safety of disposing of nuclear or toxic waste in an underground repository crucially depends upon an understanding of contaminant transport in the geosphere. In hard rock, such as granite, slate and consolidated claystone, essentially all water flow takes place through a network of fractures. In order to predict the transport of dissolved contaminants through the rock that serves as a natural barrier to migration, it is necessary to take into account the geometry of the fracture network and the transport properties of the individual fractures, including the interactions between the fluid in the fracture and in the rock matrix. The geometry can be derived only from large-scale investigations (e.g., Langer et al. 1989; Wang 1990; McKay et al. 1993a-c); the transport properties of the individual fractures can be determined from either in situ or laboratory tests (e.g., Witherspoon et al. 1980; Mull & Pfingsten 1991; Cliffe et al. 1993; Persoff & Pruess 1995).

Flow and transport phenomena in individual fractures have been investigated extensively by a number of researchers (e.g., Louis 1967; Wittke 1984; Frick et al. 1992; Puscheck 1994). The evidence from these experiments is that water flow in jointed rocks is unevenly distributed. Most of the water flow occurs over a small proportion of the fracture surface ("channeling").

Despite the complexities of real fractures, it is necessary to develop an adequate schematization to describe their flow and transport properties. Advection and dispersion processes in the fracture plane are considered in this paper, together with the diffusion of dissolved tracer into the stagnant pore water of the rock matrix. This "matrix diffusion" has been demonstrated to be a significant mechanism for retarding the migration of dissolved contaminants (Grisack & Pickens 1980; Tang et al. 1981; Heer & Hadermann 1994).

The flow and transport modules of the ROCKFLOW computer code (Zielke et al. 1994) has been used to model matrix diffusion. ROCKFLOW is a 3-D finite-element modeling program package for simulating flow and transport processes in porous and fractured-porous media. It was developed at the Institute for Fluid Mechanics and Computer Applications in Civil Engineering of the University of Hannover. Flow and material transport are represented by a continuity equation, Darcy's law and a transport equation. The transport equation incorporates advection, hydrodynamic dispersion and diffusion (after Scheidegger), a decay law, and linear isothermal sorption. Isoparametric 2-node line-segment elements, 4-node planar elements, and 8-node spatial elements are employed in the numerical realization. Coupling these elements of different dimensions makes it possible to model well-defined fractures or fault zones in a porous rock matrix.

Since publication of the initial modeling results, (Lege 1995; Lege et al. 1996), further discussions and new measurement results made recalulation with different parameter combinations necessary. On the one hand, the new results proved that the old ones were correct to some extent. On the other hand, improvements of the fit of calculated breakthrough curves to measured results are achieved mainly through a more exact determination of the effective diffusivity by Maier and Döhröfer (1994), which were not available at the time of the first papers. Additionally, the transport velocities in the fracture seem to be even higher than previously assumed. Thus, the new value calculated for the spreading of the
contamination plume at the Mönchehagen site is slightly larger now. However, the main conclusions of the our earlier publications are confirmed. The improved results are presented in this paper.

2 Mönchehagen site

The abandoned hazardous waste disposal site at Mönchehagen is about 60 km west of Hannover in Lower Saxony, Germany. Extensive sets of data covering more than a decade of groundwater monitoring are available. The settling basins of the Mönchehagen landfill are in Lower Cretaceous silt-claystone that serves as a natural barrier to migration. The rock is more than 100 m thick, comparatively well consolidated and highly fractured (Fig. 1). It is covered by thin (<2 m), unconsolidated Quaternary deposits.

The hydraulic properties of the ground have been determined in several extensive studies (Dörhöfer et al. 1994). The recorded groundwater contamination shows a discrepancy between the measured extension of the leachate plume and the spreading of the solutes expected on the basis of groundwater velocity (Fig. 2). Assuming reasonable hydraulic parameters, the measured spreading of the plume could not be matched by simple advection-dispersion models. All parameter combinations yielded much too large spreading velocities. To determine the involved retardation effects, different forced-gradient multi-tracer tests were carried out in an upstream test area. Diffusion experiments were performed with rock samples from the site in diffusion cells (Maier & Dörhöfer 1994).

Afterwards, the tracer tests were modeled numerically to determine the different conductivities of fractures and rock matrix separately in order to investigate the influence of matrix diffusion on the transport process.

The coupled effect of matrix diffusion and sorption is an important property for characterizing fractured claystone as a geological barrier for waste disposal sites.

3 Application of standard models

Models of flow and solute transport in groundwater often assume a homogeneous porous medium. The characteristics of the model area are described with "equivalent" parameters (e.g., equivalent conductivity or equivalent porosity). These parameters frequently do not give a good approximation of the conditions actually found in nature. In fact sometimes the predictions are totally wrong.

In a first approach, an equivalent porous medium is used to simulate the contaminant transport during the last 20 years from leachate from the abandoned waste disposal site at Mönchehagen. The model is a 2-D transport model, which is based on a steady-state flow field. The applied transport equation can be represented as follows:

\[
\frac{\partial c}{\partial t} = \text{div} D \text{grad} c + \frac{v_D}{n_e} \text{grad} c
\]

(1)
Table 1. Parameters of numerical 2-D model

<table>
<thead>
<tr>
<th>$K_T$</th>
<th>$\eta$</th>
<th>$c_L$</th>
<th>$c_T$</th>
<th>$i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1.5 \times 10^7$ m/s</td>
<td>10 %</td>
<td>8.0 m</td>
<td>1.6 m</td>
<td>0.004</td>
</tr>
</tbody>
</table>

The model parameters in Table 1 were taken from the on-site and laboratory studies. Equivalent hydraulic conductivity and porosity values were used for the study area as a whole. The values represent the lowest plausible parameter values for the site. Any lower value for the effective porosity $\eta$ or conductivity $K_T$ would be too far from the measured physical reality.

![Diagram](Muenchehagen 2D Standortmodell)

The contamination plume already reaches the model boundary after the simulation of a ten year period (Fig. 3). The front of the contamination plume in the model migrates obviously faster than the measured concentrations in Figure 2. Thus, it must be concluded that the porous medium approach yields a completely incorrect concentration distribution.

Retardation caused by sorption might be taken into account in the transport equation. Introduction of a retardation factor into equation (1) yields

$$ R \frac{\partial c}{\partial t} = \text{div} D \text{grad} c - \frac{v_D}{\eta} \text{grad} c $$

But if groundwater flow takes place only in the fractures, would there be enough sorption capacity on the fracture surfaces? And how do we explain that even substances like nitrate and bromide that cannot be sorbed show delayed spreading velocities and in breakthrough curves with a long tail? The latter cannot be explained by retardation as a result of sorption. Thus, the transport should not be modeled with a retardation factor that obscures physical reality. The physical process involved is diffusion into the stagnant pore water of the rock matrix. And since diffusion is a well understood...
process that is described by measurable diffusion coefficients, this process must be explicitly taken into account.

4 Matrix diffusion in fractured claystone

Matching of the history of the last 20 years with the results of the equivalent porous medium model for contaminant transport shows that this model cannot reflect the measured reality. The investigations of Maier and Dörhöfer (1994) show that in a fractured geological barrier, groundwater flow takes place almost only in the fractures. The flow velocity in the rock matrix is negligible. Thus, different transport mechanisms must be considered for fractures and for the matrix.

The dissolved contaminant is transported advectively with the actual groundwater flow in the fractures; the contaminant diffuses into the stagnant pore water of the matrix (Fig. 4). The porosity and the tortuosity τ of the pore space reduces the molecular diffusion coefficient $D_m$ to the value of the effective diffusion coefficient $D^*$. A relation between $D_m$ and $D^*$ is given by Berner (1971)

$$D^* = D_m \cdot \frac{n}{\tau^2}$$

where $n$ is the rock porosity. According to the definition of Bear and Bachmat (1990), the tortuosity $\tau$ is always larger than 1. Consequently, $D^*$ is always smaller than $D_m$.

In this study, the effective diffusivities were measured with the through-diffusion technique. This procedure, which has been described in detail by Bradbury and Green (1985), involves placing a sample of rock between a solution containing a groundwater tracer that is not sorbed and a solution in which the tracer is initially absent in a way that no hydraulic gradient exists across the sample. Thus, transport of the substance through the water-filled pores of the rock takes place solely by molecular diffusion. The increase in concentration of the tracer on the side of the sample that initially contains no tracer is monitored until a steady-state diffusive flux is attained. At this stage, the effective diffusion coefficient of the porous rock matrix may be calculated.

5 The "brush model" approach

A new model which combines 2-D finite elements for the fracture and 1-D finite elements for the matrix was developed for calculation of solute transport involving matrix diffusion. This model is called the "brush model". The advantage of the new concept is that the main physical characteristics of the different transport mechanisms in the fracture and the rock matrix are
maintained.

In this approach, gravitational effects are neglected and thus the schematization of nature yields a system of subhorizontal fractures with an attached rock matrix. If it is assumed that the dissolved substances are transported with the same velocity over the entire rock column, a model of one horizontal fracture is representative for the entire system (Fig. 5). Due to symmetry considerations, only one half of the system is modeled. The fracture plane is discretized by 2-D quadrilateral finite elements in the x,y plane with Cartesian coordinates. Advective-dispersive transport without any retardation is assumed. The transport of a substance that is not sorbed through a fracture is modeled using the following 2-D advection-dispersion equation:

$$\frac{\partial c}{\partial t} = D_L \frac{\partial^2 c}{\partial x^2} + D_T \frac{\partial^2 c}{\partial y^2} + \frac{v_D}{n_f} \frac{\partial c}{\partial x},$$

(4)

where

c: solute concentration [mg/L],
t: time [s],
v_D: Darcy velocity [m/s],
n_f: effective porosity of the fracture plane,
D_L: $= \alpha_L * v_D/n_e + D_m$: Scheidegger's longitudinal dispersion coefficient [m$^2$/s],
D_T: $= \alpha_T * v_D/n_e + D_m$: Scheidegger's transversal dispersion coefficient [m$^2$/s],
D_m: molecular diffusivity [m$^2$/s],
$\alpha_L$: longitudinal dispersivity [m],
$\alpha_T$: transversal dispersivity [m].

In the processes described in this paper, the tracer velocity, v_D/n_e, is sufficiently high that the term D_m does not contribute significantly to the total dispersion coefficients, D_L and D_T.

Darcy's law is applied for the flow velocity in the fracture, in accordance with various authors (e.g., Bräuer et al. 1989).

The rock matrix of the claystone is discretized using 1-D elements. The elements are positioned parallel to the z-axis and coupled to the 2-D elements of the fracture through common nodes. A chain of 1-D elements is attached to every node of the 2-D mesh (Fig. 6). The cross-sectional area of the 1-D elements is equivalent to the area surrounding the nodes of the corresponding 2-D mesh. Concentration in the rock matrix satisfies the 1-D diffusion equation

$$\frac{\partial c}{\partial t} = D^* \frac{\partial^2 c}{\partial x^2},$$

(5)

where $D^*$ is the effective diffusion coefficient. This schematization uses the combination of elements with different dimensions that is implemented in the applied computer code ROCKFLOW.

Of course it is also possible to model the rock matrix with 3-D elements. The Neumann number,
\[ F_0 = \frac{D^*}{\Delta t} \leq 0.5, \]  

(6)

a necessary stability criterion for modeling diffusive transport, dictates that the element width in the z-direction be much smaller (by a factor of 10 to 50) than in the x,y plane. A representative element width \( \Delta l \) was used in equation (6). \( \Delta t \) is the time step size. The computations showed that \( F_0 \) must be between 0.5 and 0.05 for correct numerical results. Thus, the 3-D elements are very distorted. Moreover, computation time is 10 times larger with the 3-D-element model than with the brush model. Additionally, the results of the 3-D model and the brush model do not show significant differences. This is due to the small diffusive transport velocity in the rock matrix compared to the advective transport velocity in the fracture. Consequently, diffusion parallel to the fracture plane can be neglected. However, if the two transport velocities are of the same order of magnitude, it can be shown that the element dimensionality influences the modeled lateral diffusion (Kolditz 1995).

6 Calibration and testing of the brush model

6.1 Set up of tracer test site

Ten observation wells were drilled in an area upstream from the abandoned waste disposal site (Fig. 7). Various hydraulic well tests and multicomponent tracer tests were carried out. The brush model was calibrated and tested using the data from tracer tests Ib and II, respectively. During the tracer experiments, a steady-state radial convergent flow field was maintained by constant withdrawal of 0.3 m³/h from well 8. In experiment Ib, microparticles (\( \phi = 1 \mu m \)) and sodium nitrate were injected simultaneously in a single pulse in well 6. Wells 6 and 8 are 6 m apart. In experiment II microparticles (\( \phi = 2 \mu m \)) and lithium bromide were injected over a period of 14 days followed by injection of pure water for 200 days. The injection and extraction wells are 18 m apart. The polystyrene microparticles move only in the fractures. They cannot diffuse into the matrix because their diameter is larger than the diameter of the claystone pores. Sodium nitrate and lithium bromide diffuse into the stagnant pore water of the rock matrix. They are tracers that are negligibly sorbed.

Tracer concentration measurements were made in observation well 8. The concentration values show an early breakthrough of the microparticles in both experiments. The breakthrough curves for nitrate and bromide are delayed and show long tails. The experimental setup and the results are described in more detail by Dörhöfer and Maier (1993).

6.2 Modeling of the tracer experiments

Tracer test IB

The brush model was initially fitted to the data from tracer experiment Ib (Lege 1995). The calculated breakthrough values fit the measured ones quite well, but normalization of the curves was necessary. The values are given in Table 2. A closer look, especially at the values for the dispersivities \( \alpha_{LP} \) and \( \alpha_{TF} \) in the fractures, shows that these values are almost certainly too large. In tracer tests in the
field the dispersivities are generally assumed to be 0.1 – 0.2 of the length of the flow path (e.g., Ptak & Teutsch 1994). For experiment Ib, Maier and Dörhöfer (1994) used a 1-D model (Rowe & Booker 1991) with $\alpha_L = 1$ m to fit the test data. With a lower $\alpha_L$ value, the peak of the calculated breakthrough curve would appear later and the flanks would be steeper (cf. tracer test II). Thus, a higher conductivity value would be necessary to fit the experimental data. A sensitivity analysis of $\alpha_L$ and $\alpha_T$ in the modeling of experiment II supports this assumption. Only parameter variations of tracer test II are shown in this paper.

<table>
<thead>
<tr>
<th>numerical model of tracer tests Ib and II: fixed parameters</th>
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<tbody>
<tr>
<td>$K_{sp}$ [m/s]</td>
</tr>
<tr>
<td>----------------</td>
</tr>
<tr>
<td>microparticles (test Ib)</td>
</tr>
<tr>
<td>bromide (test Ib)</td>
</tr>
<tr>
<td>microparticles (test II)</td>
</tr>
<tr>
<td>bromide (test II)</td>
</tr>
</tbody>
</table>

sensitivity analysis (tracer test II): variable parameters

- model A
- model B
- model C
- model D
- model E

accessible diffusion space (length of bristles): 2.8 cm

$D^* = 9.4\times10^{-10}$ m²/s 2.5×10^{-10} m²/s 1.4×10^{-10}$ m²/s 1.0×10^{-10}$ m²/s 2.5×10^{-10}$ m²/s

**Tracer test II**

As depicted in Figure 8 the choice of $\alpha_L = 8$ m and $\alpha_T = 1.6$ m is correct in this case. Breakthrough curves of the microparticle experiments were calculated with different dispersivities. The curves with smaller dispersivities show a later peak and – more important – a steeper drop after the peak concentration is reached. From the microparticle experiment, the conductivity $K_{sp}$ of the fracture is $5.9\times10^{-3}$ m/s. Thus, the measured data is matches best with $\alpha_L = 8$ m (Table 2).

The bromide test was modeled with the advective-dispersive transport properties of the fracture – the handle of the brush. Additionally, chains of 1-D diffusion elements – the bristles of the brush – are attached to

**Figure 8.** Normalized breakthrough curves of tracer test II (microparticle experiment) showing the results of variation of dispersivity (cf. Table 2).
every node of the discretized fracture plane (cf. Fig. 6). The effective diffusion coefficient $D'$ and the accessible diffusion space (i.e., the length of the 1-D element chains, which is a measure of the distance between fractures) were varied in a sensitivity analysis. Maier and Döhröfer (1994) used a diffusivity $D = 9.4 \times 10^{-10} \text{ m}^2/\text{s}$ and a porosity of 14% for the claystone matrix, and tortuosity is not mentioned. Thus, the tortuosity was set to 1 and equation (3) yields $D' = 1.3 \times 10^{-10} \text{ m}^2/\text{s}$.

The best fit was obtained with $D' = 1.0 \times 10^{-10} \text{ m}^2/\text{s}$. The results of different models are depicted in Figure 9: Larger diffusivity values result in later peak arrival time and lower peak concentrations. In addition, the tails are longer. An explanation for this is that with higher diffusivities, more of the dissolved substance diffuses out of the fracture into the matrix. The substance travels faster and thus deeper into the rock matrix. Thus, the peak concentration of the breakthrough curve is lower. Consequently, the backward diffusion from the rock matrix into the fracture after the concentration peak passed is slower and the tails are longer. However, the results of this study improve the outcome of Lege (1995), where $D' = 2.5 \times 10^{-10} \text{ m}^2/\text{s}$ was assumed on the basis of the initial evaluation of diffusion experiments. Unfortunately the more exact results of Maier and Döhröfer (1994) were not known at that time.

It is of interest to note that extending the accessible diffusion space does not significantly influence the breakthrough curve (Fig. 9; models B and E).

It may be concluded that on the time scale of these tracer tests, diffusivity is a more sensitive parameter than the distance between the fractures. In the following section it is shown that larger time scales yield different results.

### 7 Model of the site

**History matching**

Since the brush model was calibrated a site model has been developed. The conceptual model is the same as the one for the tracer tests:

![Figure 10. Calculated contamination plume after 20 years of leachate infiltration from the old settling basin into the groundwater in the geological barrier at the Münchhagen site.](image)
Flow and transport in the fractures of the claystone are modeled in a 2-D horizontal plane discretized with 2-D elements (the brush handle). Chains of 1D elements are attached to each node of the 2-D plane perpendicular to the plane (the bristles of the brush). They represent the rock matrix. The parameters of the site model are summarized in Table 3.

The calculated contamination plume of the older, westernmost settling basin of the site is depicted in Figures 9 and 10. Comparison with Figure 2 shows good correspondence between the calculated and the measured plumes. After 20 years of leachate infiltration into the groundwater, the plume had spread only 55 m downstream. The concentration at the front of the plume is four orders of magnitude less than the values of the boundary conditions. Very high pollution was calculated only in the immediate neighborhood of the old settling basins in agreement with the measurements in the field. The calculations show good agreement with the observed data. Thus it is correct to conclude that in spite of considerable generalization, the brush model can yield good results for transport calculations in fractured systems with matrix diffusion as a major retardation factor.

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7.2 Predictive modeling and sensitivity analysis

Various parameter combinations were computed for sensitivity analysis and predictive modeling (Table 3). The calculated concentrations in downstream observation wells (42 m and 78 m from the edge of the landfill) are shown in Figures 11 and 12. A period of 100 years was simulated. On this time scale,
the distance between the fractures, the accessible diffusion space, is a more sensitive parameter than the diffusion coefficient. The curves of models II and III, as well as the ones of model I and IV, are fairly close to each other. The sole difference between models I – V is the diffusion coefficient and the accessible diffusion space. The fracture conductivity $K_f$ was derived from tracer test IB. The time scale is apparently large enough to show the influence of saturation of the stagnant pore water with contaminants: If the same concentration as in the mobile water of the fracture is reached in the pores, matrix diffusion ceases to delay the spreading process. This was not observed at the short-term tracer tests.

If the conductivity of the fracture is further enlarged (closer to the very high values of tracer test II), there might be not sufficient time for the dissolved substance to diffuse into the matrix. The downstream concentration rises faster (compare the curves of models IV and V). However, after 100 years in an observation well 78 m downstream, the concentrations are still much lower ($\leq 6\%$) than the initial values.

Table 3. Initial and boundary conditions and parameters of the site model

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<thead>
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<th>Initial and boundary conditions</th>
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<tbody>
<tr>
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<td>downstream boundary (SW)</td>
<td>$c_B$: free</td>
<td>streamline boundaries (NW, SE)</td>
<td>$c_B$: free</td>
</tr>
<tr>
<td>Rim of landfill</td>
<td>$c_B = 100%$</td>
<td>region outside of landfill</td>
<td>$c_I = 0%$</td>
<td>region inside of landfill</td>
<td>$c_I = 100%$</td>
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</table>

Parameters of the site model

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<thead>
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<th>parameter</th>
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<th>model II</th>
<th>model III</th>
<th>model IV</th>
<th>model V</th>
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<td>fracture $n_{fr}$</td>
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<td></td>
<td>$1 \times 10^{-3}$ m/s</td>
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<td>$2.5 \times 10^{-10}$ m²/s</td>
<td>$9.4 \times 10^{-10}$ m²/s</td>
<td>$1.4 \times 10^{-10}$ m²/s</td>
<td>$1.4 \times 10^{-10}$ m²/s</td>
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<td>longitudinal dispersivity $\alpha_L$ (fracture)</td>
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<td>transversal dispersivity $\alpha_T$ (fracture)</td>
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<td>hydraulic gradient $i$</td>
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<td>half fracture aperture $b$</td>
<td>$0.0001$ m = $100$ µm</td>
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<tr>
<td>half distance between fractures</td>
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<td>$0.195$ m</td>
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8 Summary and Conclusions

With the brush model approach presented here, diffusion from the mobile groundwater in fractures into the stagnant pore water of the rock matrix can be taken into account elegantly. Especially in argillaceous formations, matrix diffusion very effectively retards the spreading of contaminants. The brush model approach assumes that the immobile pore water of the rock matrix is connected with the flowing groundwater in the fractures. If contaminated water flows through the fractures, a diffusion process is initiated due to the concentration gradient between the fracture and the rock matrix.

In order to model the combined transport processes of advection and dispersion in the fractures and diffusion in the matrix with the finite element method, a new conceptual model has been presented in this paper. The fracture plane is represented by 2-D quadrilateral advection-dispersion elements – the brush handle. Attached to each node of the 2-D mesh are chains of 1-D diffusion elements perpendicular to the fracture plane.

Comparisons of modeling results with field data from two different multicomponent tracer tests proved the applicability of the approach to real world problems. This finding is further supported by a history matching of the current groundwater pollution situation at an abandoned waste disposal site in fractured claystone.

The new approach has the following advantages:

- Diffusion coefficients from laboratory experiments can be transferred directly into the modeling of field observations.
- The finite length of the diffusion elements can be used to take saturation effects into account, or in other words the limited diffusion capacity of the rock matrix.
- Calculation of the backward diffusion after peak concentration in the fracture can be modeled.
- The approach is an elegant and simple conceptualization of a rather complex geological reality.

To summarize, one can conclude for the understanding of transport processes in a fractured argillaceous formation it is important to realize that

- hydraulics are not sufficient for the characterization of argillaceous formation as a geological barrier for radioactive waste disposal;
- the physical and chemical interactions between the contaminant and the rock are very important;
- for long time periods (years), the distance between fractures is a more sensitive parameter for contaminant retention than the diffusion coefficient. The opposite is true for short time scales (weeks to months).

9 References


Fault Geometry and Groundwater Flow
in an Alpine Marl Formation

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Abstract

On the basis of an extensive drilling campaign (7 boreholes with depths between 430 and 1870 m below surface), faults have been identified to be the dominant water flowpaths in the marl formations at Nagra's proposed repository site at Wellenberg (Helvetic nappes of the Central Swiss Alps). Each fault consists of one or more horizons of fault gouge/fault breccia (thickness: 1 mm - few decimeters) surrounded by a fault damage zone (thickness: centimeters - meters) in the adjacent wallrock. Flow takes place either along the fault gouge horizons or within the microfractures of the damaged zone.

Transmissivities of faults decrease with depth from ca. $10^{-7}$ m²/s in the uppermost 200 m to $<10^{-9}$ m²/s below ca. 500 m. Even at a given depth below ground, the transmissivity varies over 3 orders of magnitude, indicating structural and hydrogeologic heterogeneity of the faults. At shallow levels, a large part of all faults identified in the core material have transmissivities above the detection limit of the fluid logging tool (ca. $10^{-9}$ m²/s) and thus were identified as inflow points. In contrast, only a fraction of all existing faults are identified as inflow points at depths greater than ca. 300 m, indicating that flow is channelled and large areas within faults have transmissivities below detection. Geologically, permeable faults cannot be distinguished from those that do not represent inflow points. Thus while the frequency and the geometry of faults do not vary significantly with depth, their transmissivity decreases substantially. The larger transmissivities at shallow depths are best explained by an increase of the fracture apertures and of the connectivity due to the decrease of the overburden by erosion and/or postglacial decompression. The enhanced fracture apertures at shallow depths are also indicated by much lower shear-wave velocities when compared to the deeper parts of the boreholes.

The majority of all faults lies concordantly within the most argillaceous strata of the formation and dip moderately to the NW. More competent limestone strata are generally cross cut discordantly by more steeply dipping normal fault segments. The dominant part of the flowpath through faults is situated within the argillaceous strata.
Flow modelling using a fracture network approach requires information on fault transmissivity, frequency, orientation, internal structure (heterogeneity of flow) and fault length. All these parameters can be derived on the basis of the geologic and hydrogeologic borehole database, with the exception of fault length where large uncertainties remain. Some information on fault length and internal structure can be obtained from surface outcrops with maximum observation windows in the order of 10 m x 10 m. The problem remains that the fault size often exceeds that of the outcrop, so that outcrop information must be extrapolated (scaled up) to properly represent faults on the scale of the block model (cube with 500 m edges). Remaining uncertainties must be handled by parameter variations in the model runs, and additional information from hydrochemistry must be integrated for further constraints (such as consistency of geochemically derived groundwater residence times and hydrogeologically modelled transit times).
Fault-Controlled Groundwater Flow in Mudrocks at Down Ampney, UK: Geochemical Evidence

R Metcalfe, S Reeder, M R Cave, K A Green, D C Entwisle and J R Davis

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Keyworth, Nottingham
England, NG12 5GG-UK

Abstract

The hydrochemistry of a site at Down Ampney in Gloucestershire, UK has been studied by means of conventional groundwater sampling and analysis of pore-waters collected from drillcore samples of mudrocks by mechanical squeezing. The geological succession at the site consists of a thin layer of terrace gravels overlying Oxford Clay, which in turn overlie the Kellaways Beds and the main limestones of the Jurassic. Samples were taken from boreholes up to 100m deep across an area straddling a prominent east-west trending fault.

The pore-waters are mainly Na-Cl dominated, except in the fault zone where Na-SO$_4$ waters dominate. Inverse correlations between pH and SO$_4$ indicate that pH (and, in turn, carbonate solubility and hence cation concentrations) are probably controlled by oxidation of sulphides (probably pyrite) by oxidising meteoric water. The dominance of Na-SO$_4$ rather than Ca-SO$_4$ rich waters in the faulted clay, as might be expected from sulphide oxidation, may be explained by cation exchange with Na-bearing phases or by the evolution of a more saline, oxidising groundwater from depth.

The evidence from the geochemical data, including stable isotope compositions, suggests that modern meteoric water penetrates downwards through the mudrocks, diluting groundwaters and pore-waters that are present already, with the major fault acting as a short-circuit to groundwater flow. The hydrochemistry of the site may also be controlled by the influx of oxidising Na-SO$_4$ rich waters, possibly originating at depth in the Kellaways Beds, being channelled up through the fault to shallower depths.

The study demonstrates that geochemical data derived from a combination of pore-waters and groundwaters can be valuable for resolving geologically recent groundwater flow in mudrocks.

Introduction

Evidence for fault-controlled, paleo-fluid flow in mudrocks is often provided from features of the rock mass such as sedimentary de-watering structures and mineral veins. The detection of geologically recent, fault-controlled groundwater flow, however, is rather more difficult because: (i) readily observable structural or mineralogical evidence is often lacking;
(ii) low and/or perturbed (by physico-chemical effects such as osmosis) groundwater flows render groundwater flow directions and magnitudes difficult, or even impossible, to deduce from groundwater heads; and (iii) the temporal relationships between physical evidence for fluid flow and modern groundwaters are usually unclear (e.g. it is often difficult to prove whether a mineral vein precipitated from modern groundwater or some paleo-fluid).

Geochemical evidence from modern groundwaters and pore-waters may, therefore, provide the only direct evidence for geologically recent, fault-controlled fluid flow. This paper describes a pilot study applying hydrogeological techniques to detect groundwater flow in faulted mudrock at the Down Ampney Fault Research Site in Gloucestershire, UK (Figure 1).

![Map of the United Kingdom showing the location of Down Ampney](image)

**Figure 1** The location of the Down Ampney site

### The Study Site

The geology of the site consists of a sequence ranging from the Oxford Clay to the Taynton Stone. A thin layer of terrace gravels overlies up to 75m of Oxford Clay which is variable in lithology, but comprises mostly calcareous mudstones with silty/shelly horizons and occasional thin limestone bands. These mudrocks in turn overlie the sandy Kellaways
Beds, the main limestones of the Jurassic succession. These consist of a 10-12m thick upper layer of fine-grained sands and sandy silts, and a 2-3m lower layer of silty mudstone. The Cornbrash, beneath the Kellaways Beds, consists of fine sparites and thinner marls and overlies a succession of calcareous lithologies comprising the Forest Marble, White Limestone, Hampen Marly Beds and Taynton Stone.

Samples were taken from boreholes across an area straddling a prominent east-west trending fault with a northerly downthrow of c. 50m (Figure 2). The investigation focused on three boreholes, up to c. 100m deep: DA1, upthrow and south of the fault; DA11, which penetrates the fault and DA2, downthrow and north of the fault (Figure 3). DA1 and DA2 are positioned several tens of metres away from the fault.

![Figure 2](image)

**Figure 2.** Plan of the borehole array at the Down Ampney site

**Techniques**

The measurement of the chemical composition of waters obtained from aquifers is relatively straightforward, but it is more difficult in mudrocks because of their low permeability. To compliment the relatively few groundwater samples obtained by pumping
with a submersible pump or by air-lifting, it is necessary therefore to extract pore-waters from core samples by mechanical squeezing (Entwisle and Reeder, 1993; Reeder and Entwisle, 1995).

![Diagram of borehole locations and geology of the site](image)

**Figure 3.** Borehole locations and geology of the site

Sample core was extracted from the borehole in stainless steel liners and the ends sealed in wax to preserve their moisture content. The core samples were recovered for testing by cutting the liners vertically and reaming off the outermost section of core using a soil lathe. Pore-waters were then extracted from the core by squeezing in constant temperature (10°C) compression cells specially designed at the British Geological Survey. Small initial stresses of 1-3 MPa were initially applied to bed the sample into the squeezing cell and expel any air. Stress was then increased until pore-water was extracted and the stress maintained, or if necessary progressively increased (to a maximum 65 MPa), until about 20ml of pore-water was obtained.

Groundwaters and pore-waters were analysed using conventional analytical techniques. Major and trace cations were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) using a Perkin Elmer Plasma II emission spectrometer. Chloride, sulphate, nitrate, nitrite, phosphate and bromide were determined by ion chromatography using a Dionex 2000i ion chromatograph. Fluoride was determined using an Orion 24-09 ion selective electrode. Total organic and inorganic carbon were analysed on a Shimadzu TOC 500 analyser. Carbonate and bicarbonate alkalinity were determined by titration against 0.01M sulphuric acid using a Radiometer VIT 90 automated titration system.
Results

Comprehensive results for all samples collected as part of the study are given in Metcalfe et al. (1990).

Total cation concentrations in the pore-waters range from 9.23 to 178 meq l⁻¹. Most of the waters are Na-Cl dominated, except in the fault zone where all but one of the waters are Na-SO₄ dominated. Na-SO₄ waters also occur in the Kellaways Beds within borehole DA1 (Figure 4).

Figure 4. Trilinear plot showing major element composition of groundwater and pore-water samples from Down Ampney

Figure 5. Plots of Na/Ca mass ratio versus depth for Down Ampney boreholes DA2 and DA11
Cation Na/Cl ratios generally increase towards the fault in borehole DA11. Away from the fault, in borehole DA2, there is less systematic variation. The maximum Na/Ca ratios occur in the fault zone (Figure 5).

Both near the fault and remote from the fault, pH correlates positively with alkalinity and SO₄ in some places, but negatively with others (Figures 6 and 7).

**Figure 6.** Plots of pH, alkalinity and sulphate versus depth for Down Ampney borehole DA11 pore-waters

**Figure 7.** Plots of pH, alkalinity and sulphate versus depth for Down Ampney borehole DA2 pore-waters

Salinities broadly increase downwards across the mudrocks, but then decrease in the Kellaways Beds and near the fault (Figure 8).

The stable isotopic compositions of the pore-waters lie on the meteoric water line. Deeper waters are depleted in heavy isotopes relative to the shallower waters (Figure 9). Values range from δ¹⁸O_MOW c. -6.5‰ to -8.0‰ and δ²H_MOW c. -44‰ to -55‰.

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Figure 8. Plots of Cl versus depth for Down Ampney borehole DA1, DA2 and DA11.

Figure 9. Plots of $\delta^{18}$O versus $\delta^2$H and $\delta^{18}$O versus depth for Down Ampney boreholes.
Discussion

Hydrochemical data obtained from both groundwaters and pore-waters are broadly similar (Figure 4), indicating that the pore-water chemistry is not significantly perturbed by the squeezing process. This observation has been confirmed by other studies, e.g. Bath et al. (1989a) and Gautschi et al. (1993). The results obtained do need to be treated with some caution, however, because of the low density of sampling.

Salinity concentrations in the pore-waters (identified by Cl, Br and F concentrations) increase with increasing depth. In boreholes remote from the fault, however, the salinity gradients are approximately twice as great as those observed in the borehole intersecting the fault. This variation in salinity gradient probably reflects contrasts in permeability, with the fault, and possibly the Kellaways Beds, channeling dilute water.

Negative correlations observed between pH and sulphate (and pH and alkalinity) occur mainly in the fault zone and below relatively permeable layers. This indicates that pH is controlled largely by sulphide (probably pyrite) oxidation in the fault zone arising from fault-controlled flow of oxidising meteoric water. The sulphate oxidation leads to decreases in pH causing carbonate mineral dissolution and increasing cation concentrations. This is expected to cause Ca-SO\textsubscript{4} dominated waters, not Na-SO\textsubscript{4} dominated waters as observed in the fault zone and the Kellaways Beds of borehole DA1. The Na-enriched compositions may be explained by displacement of Na by cation exchange with Ca from Na-bearing phases of the clay, occurring after the initial pyrite oxidation and subsequent calcium carbonate dissolution. Alternatively, the Na-SO\textsubscript{4} could have evolved from a more saline, oxidising groundwater, possibly originating at depth in the Kellaways Beds.

The isotopic data indicate that the waters are meteoric. Decreases in stable isotopic compositions with depth are consistent with more recent recharge of shallower waters compared to deeper waters. The isotopic values given by the deepest samples are typical of depleted meteoric waters involved in Pleistocene recharge in the colder climate prevailing during or just after the last glacial maximum, around 20000 years ago. An alternative explanation for the isotopic shift is near-surface isotopic fractionation, similar to those observed by Bath et al. (1989b), although this is considered less likely than the paleoclimatic variation model.

Conclusions

The overall model, presented in Figure 10, is that modern meteoric water penetrates downwards through the mudrocks throughout the site, diluting groundwaters and pore-waters that are present already. The major fault acts to short-circuit groundwater flow, however, and flushing of pre-existing water has been particularly vigorous along this structure. The hydrochemistry of the site may also be controlled by the influx of oxidising Na-SO\textsubscript{4} rich waters, possibly originating at depth in the Kellaways Beds, being channelled up through the fault to shallower depths.

The study demonstrates that geochemical data derived from a combination of pore-waters and groundwaters can be valuable for resolving geologically recent groundwater flow in mudrocks.
Figure 10. Schematic conceptual model for the hydrogeology of the Down Ampney Fault Research site based upon the hydrochemical data

Acknowledgements

The work reported in this paper was undertaken as part of a research programme funded by Her Majesty’s Inspectorate of Pollution and the Commission of the European Communities.

Dr A H Bath is thanked for reviewing the original interpretation and suggesting many improvements to it.

References


Links between Water Transfer and Fracturation in a Clay Formation (Tournemire, France): Isotopic Approach.

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Jean-Yves Boisson
Institut de Protection et de Sûreté Nucléaire, France

Abstract

An isotope hydrology study of interstitial water and water flowing from faults and fractures is being performed on a claystone massif (Tournemire, France). $^{18}$O, $^2$H and $^3$H contents allow to distinguish different origins of water: (i) porewater of the non fractured zone could be meteoric water, possibly marked by a recharge in cooler conditions than present. (ii) porewater of the fractured zone would have been affected by a secondary process of enrichment in heavy isotopes (iii) water flowing from the major fault shows a recent meteoric origin, being connected to the over and underlaying karstic aquifers (iv) water flowing from the fractured zone shows either a mixing between recent water and porewater, or a relatively long transit time.
Introduction

Isotope hydrology is one of the tools that can be used to understand the water transfers in very low permeability media such as clay formations. In the Tournemire site, which is studied by the IPSN since 1988, isotopic investigations are being performed. They concern porewater ($^{18}$O, $^3$H) and water flowing from faults which affect the massif ($^{18}$O, $^3$H, $^3$H).

Geological view

The claystone of Tournemire, southern France (Fig. 1), is a Toarcian formation. The mineralogy consists of clay minerals (Kaolinite, Illite-montmorillonite) and Mica (40%), of Quartz (20%), Calcite (20%) and other phases: Dolomite, Pyrite, Feldspat and Siderite [1]. The water content of the claystone is lower than 5% by wet weight, and the permeability coefficient was estimated by several methods to be lower than $10^{-13}$ m.s$^{-1}$ [2].

This thick claystone formation (250 metres) is over and underlayed by limestones and dolomites where karstic aquifers are developed (Fig. 1).

Figure 1: Location of Tournemire (Aveyron, France) and schematic geological cross section of the site.

The structural patterns of the claystone formation are inherited from two major tectonics phases: the Pyrenean compression at Eocene (N-S), and the Oligocene extension (E-W) [1]. The massif is not affected in totality by these faults and fractures (see Boisson et al., these proceedings).
Water samples

**Interstitial water**

Eight radially boreholes (Fig. 2), covering a 16 000 m² area, were drilled from (and perpendicularly to) a tunnel which crosses the Toarcian (Fig. 1). They allow to explore a fractured zone and a non fractured zone.

**Water flowing from the faults**

Three sampling points provided enough water for isotopic analyses. The first one is the Cernon fault which crosses the tunnel (Fig.1). This fault is a major accident connecting the upper and lower karstic aquifers. Its discharge is of several litres per minute. The second and third sampling points are the two boreholes drilled in December 1994 which cross a fractured zone without reaching the upper aquifer (Fig. 2). The discharge does not exceed few ml.h⁻¹.

Claystone samples were covered just after the drilling by a thick wax layer in order to prevent isotopic exchanges between water and atmosphere, and evaporation. The method used for extracting the interstitial water was the vacuum distillation [3]

Oxygen-18 and Deuterium analyses on water were performed using respectively the methods described by Epstein (1953) [4] and Coleman (1982) [5].

**Fault waters: different origins**

**Stable isotope signature**

Stable isotope measurements of the fault waters and of the pore water in the non fractured zone are reported on a Oxygen-18 vs. Deuterium diagram (Fig. 3).

All of this samples are situated in a zone around the M.W.L. [6]. Three water types can be distinguished, corresponding to the water samples from the Cernon fault, the water samples from the fractured zone, and the interstitial water samples of the non fractured zone.

The Cernon fault is supposed to be connected to the karstic systems developed in the two aquifers. Karstic aquifers are well-known for their rapid transfers. The water flowing from the Cernon fault could be recent meteoric water. The Deuterium excess [7] shows a mediterranean influence [8] with some values near +20‰. The variation in Deuterium excess probably reflects that of respective contributions of oceanic vapour and mediterranean vapour to local precipitation.

The porewater issued from the samples collected in the non fractured zone is also marked by a meteoric signature. The Deuterium excess is close to the value measured in the water samples taken from the Cernon Fault. Nevertheless, the totality of the samples are depleted in heavy isotopes compared to the Cernon fault water. This could correspond to meteoric water recharged under mean climatic condition cooler than present ones (paleoclimatic effect).

Between the above defined zones are situated the water samples collected from the fractured area crossed by the boreholes ID 0 and ID 315. Such a position in the diagram may be due either to a mixing between water from the upper karstic aquifer and porewater or to paleoclimatic effect.
Figure 3: Stable isotope contents of interstitial water in the Tournemire claystone: non-fractured zone and of water flowing from the faults.

Tritium analyses

Tritium measurements by β-counting after electrolytic enrichment were performed on water flowing from the faults. The amount of water needed for these measurements makes the measurement on interstitial waters impossible. However, it is very likely that porewater is tritium free. Results are shown in the table 1.

<table>
<thead>
<tr>
<th>Location</th>
<th>Cernon Fault (direct connection with the lower and upper karstic aquifer)</th>
<th>Borehole ID 0 (crossing the fractured zone)</th>
<th>borehole ID 315 (crossing the fractured zone)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tritium measurement (U.T.)</td>
<td>8.2±0.5 U.T.</td>
<td>&lt;1±0.5 U.T.</td>
<td>0.9±0.5 U.T.</td>
</tr>
<tr>
<td>Residence time</td>
<td>≤ 10 years</td>
<td>≥ 50 years</td>
<td>≥ 50 years</td>
</tr>
</tbody>
</table>

Table 1: Tritium content of the water issue from the fault of the claystone massif (03/27/95).

Water flowing from the Cernon fault is clearly very recently recharged.

The water samples collected in the fractured zone show very low Tritium contents. That may, again, be explained either by a mixing between porewater (Tritium free) and recent karstic waters or by a pure radioactive decay, which would correspond to a residence time longer than 50 years. These hypotheses will be tested by measuring 14C contents of dissolved inorganic carbon.
Porewater and structural influence

Isotope contents of porewater are reported in a $\delta D$ vs. $\delta^{18}O$ diagram (Fig. 4). Distinction was made between the samples collected in the vicinity of a fracture (<2m) and those taken in a non fractured zone (distance to a fracture >2m) (Fig 2).

The presence of the fractures influences the isotopic contents of interstitial water. The samples from the fractured zone have a Deuterium excess lower than that of the karst water samples. In the diagram, they are roughly distributed around a line with a slope of 2, originating in the zone where the samples from the non fractured zone are situated. Such a low slope may be generated by an evaporation process in clayey materials [9]. The question arises if this process can occur in a medium like our argilite formation, which is assumed to be saturated. Another possible explanation consists in considering Oxygen-18 exchange with the calcite that filled, at least partially, the fractures.

![Graph showing $\delta D$ vs. $\delta^{18}O$ with lines and data points]

**Figure 4**: Stable isotope contents of interstitial water in the Tournemire claystone (fractured and non fractured zone).

Conclusion

The isotopic investigation has provided information relative to the water transfers in the Tournemire massif. The first important conclusion is that both water flowing from the faults and porewater show a meteoric origin. The Cernon fault, which is the major accident is connected to the karstic aquifers. Water flowing from this fault has a recent meteoric origin. Concerning water flowing from the fractured zone, two hypotheses could explain the isotopic contents: either a mixing between porewater and recent water, or a transit time long enough to allow a virtually complete decay of Tritium.

The porewater of the non fractured zone would mainly be of meteoric origin. Depleted in heavy isotopes by comparison to the present karstic waters, it could have been recharged under climatic conditions cooler, in average, than present. The porewater of the fractured zone could have the same meteoric origin, but would have been affected by a secondary process of enrichment in heavy isotopes, which is not fully understood for the time being.
References


Characterization of the Hydraulic Properties of the Boom Clay Formation Around the HADES Underground Research Facility

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Abstract

Laboratory experiments have shown the stress dependence of the hydrogeological parameters of plastic clay. Recent in situ hydraulic tests at CEN-SCK added evidence to this stress dependence. Since 1985, CEN-SCK is doing in situ hydrogeological characterization experiments in the Boom clay formation around the HADES underground research facility, in Mol. We obtained most of the results from the many piezometers installed either for the hydrogeological studies or for in situ radionuclide migration experiments. The scale of those experiments is rather small, i.e. tens of centimetres to a few metres. In 1993, a large scale permeability test (tens of metres), referred as the “macropiezometer test”, was initiated in the exploratory shaft and gallery located at the end of the underground research laboratory.

We finished this exploratory construction by the end of 1983. The lining is made of pervious concrete blocks, so that water is flowing in continuously. In 1993, we isolated the little shaft entrance with a glass cover, sealed with a synthetic resin, so that any hydric exchange with the main gallery located above became impossible. We tested the effectiveness of this system by measuring permanently the relative humidity at three different levels of the exploratory construction. The cumulated water inflow was monitored at regular time intervals. We compared the measured cumulative water inflow with the inflow calculated by the code PORFLOW, designed to solve multi-phase flow, heat transfer and mass transport problems in variably saturated fractured or porous media. We simulated also the pore water pressure distribution along the horizontal piezometest CP1, located at the end of the main gallery. We obtained a very good agreement between the experimental and the simulated values. The simulation results show that the hydraulic conductivity and the storage coefficient in the clay layer surrounding the exploratory construction are lower than expected. For the vertical and horizontal components of the hydraulic conductivity, we obtained values lower than 1x10^-12 m.s^-1, 2x10^-12 m.s^-1. The storage coefficient was lower than 8x10^-6 m^-1. In previous experiments in non-drained filters, we measured 2.3x10^-12 m.s^-1, 5.2x10^-12 m.s^-1 and 8.1x10^-6 m^-1 for the same parameters. This might be due to a slow consolidation process that followed the drilling of the little shaft and gallery. The local pore water pressure gradually decreased, which provoked a corresponding increase of the local effective stress.

To test the influence of the effective stress on fluid flow, we carried out water injection experiments at two different locations in the underground laboratory. The first location is a multi piezometer,
containing eleven filters, and installed vertically at the bottom of the main access shaft in 1986. The second location is a three-dimensional configuration of four horizontal multi piezometers, consisting of twenty-nine filters, and installed in 1992 from the gallery. Each filter is connected to a pore water pressure sensor. These experimental setups are respectively referred as MEGAS E4 and MEGAS E5. Figure 1 represents a global view of the location of those devices and the location of the exploratory construction.

Figure 1. Location of the exploratory construction and the MEGAS in situ experiments in the HADES underground research laboratory

The aim of the water injection experiments is to study the relationship between the hydraulic conductivity and the water injection pressure, and thus the effective stress. For a constant total stress, an increase in pore water pressure leads to a decrease in effective stress and thus to a deconsolidation of the clay.

In E4, we used a syringe pump to inject water at constant pressure in the deepest filter of the multipiezometer. The equilibrium pore water pressure in this filter is 1.75 MPa. We imposed a range of different water injection pressures (from 1.2 MPa below to 1.25 MPa above the equilibrium interstitial pressure). A steady state flow was observed, each time after about one week. The measured hydraulic conductivity as a function of the pressure difference is shown on Fig. 2. On this figure, the indicated standard deviations have been calculated on the basis of the pressure fluctuations and the precision of the measuring apparatus. The hydraulic conductivity clearly increases with increasing water pressure and thus decreasing effective stress. The hydraulic conductivity increases by 60 per cent for an overpressure ranging from zero to +0.9MPa. This increase could be an indication that there is a rearrangement of the pore structure in this pressure interval.

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We carried out the same type of experiment in a filter of E5, where the local equilibrium interstitial pressure is 1.86 MPa, and we observed the same phenomena but at a different pressure level. The hydraulic conductivity remains remarkably constant up to +1.47 MPa, and then increases sharply by 30 percent for a pressure increment of +0.1 MPa. The different range of pressure established for the occurrence of this phenomenon in E4 and E5 is most probably due to the different initial local stress state. Indeed, the oversize of the drilled borehole was larger for E4 than for E5. This allowed a larger convergence of the clay, leading to a larger deconsolidation and thus to a lower effective stress in the direct vicinity of the borehole.

![Graph showing hydraulic conductivity vs. injection overpressure](image)

**Figure 2: Hydraulic conductivity as a function of the injection pressure in E4**

It is clear that fluid flow in a dense clay layer, such as the Boom Clay at Mol, is strongly influenced by the local geomechanical conditions. A decrease of the effective stress will lead to an increase in hydraulic conductivity. Although we did not observe any hydrofracturing of the clay for water overpressures up to 1.25MPa in E4 and 1.56MPa in E5, it is not excluded that such phenomenon could occur at zero effective stress.
The Morphology and Formation Mechanisms of the Rusey Breccia, North Cornwall, U.K.

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Abstract

The Rusey breccia outcrops in the footwall of a major thrust, on the north coast of Cornwall, U.K. This breccia shows a remarkable degree of dilation, with matrix supported clasts. Up to 80% of the rock may be composed of the monomineralic quartz matrix. The clasts show a high degree of organisation, such as grading, layering and channel features. It is evident that simple brecciation cannot account for the all of the features seen in the Rusey Breccia.

The quartz mineralization identifies five phases of deformation structures. Fluid inclusion data shows the composition of the source fluid evolving with the development of the breccia, from metamorphic fluids through a phase of high salinity meteoric waters to lower salinity meteoric water. Fluid type I is restricted to the initial brecciation episode. The later fluids and structures are interpreted as being associated with continued thrusting during the Variscan orogeny, followed by late stage strike-slip and oblique extension as the orogen undergoes gravitational collapse. Although the breccia structures and fluid types show good correlation to the major structures of the area, this model for the evolution of the breccia does not explain the organised distribution of clasts within the breccia. Therefore further non-brecciation processes must be included to account for this aspect of the breccia.

The breccia was formed under unusually dilatant conditions, generating a zone of high porosity and permeability. This was then followed by intake of fluids into the brecciated zone, during ongoing deformation, initiating fluid flow processes. Seismic vibration combined with the effects of fluid flow to modify the original breccia distribution. The combination of high porosity/permeability and high fluid intake resulted in the formation of a matrix supported system, permitting unusual growth textures in the matrix.
INTRODUCTION

The Rusey breccia outcrops in the footwall of a major thrust, found on the north coast of Cornwall, U.K., (fig. 1). This breccia shows a remarkable degree of dilation, with matrix supported clasts. Up to 80% of the rock may be composed of the monomineralic quartz matrix. The clasts show a high degree of organisation, such as grading, layering and channel features, (photos 1 & 2). It is evident that simple brecciation cannot account for the all of the features seen in the Rusey Breccia.

The quartz mineralization identifies five phases of deformation structures (fig. 2, table 1). Fluid inclusion data shows the composition of the source fluid evolving with the development of the breccia (table 1, Davies, 1993). Initially the fluids were derived from formational waters and meteoric dewatering. They evolved from metamorphic fluids through a phase of high salinity meteoric waters to lower salinity meteoric water. Fluid type 1 is restricted to the initial brecciation episode, being found in the early veining within the clasts (fig. 2, table 1). Fluid type 2, also metamorphic, is found in the anhedral part of the main matrix, immediately adjacent to the clast and vein wall surfaces (fig. 2, table 1). Fluid type 3A is also found in the main matrix, in the outer euahedral crystals, but is described by Davies (1993), as a meteoric fluid of distinctly higher salinity than later fluids. Fluids 3B and 3C are found in extensional veins and high angle shear veins respectively (see fig. 2), and show steadily decreasing salinity levels. These latter fluids and structures are interpreted as being associated with late stage strike-slip and oblique extension as the orogen undergoes gravitational collapse. Although the breccia structures and fluid types show good correlation to the major structures of the area (fig. 3, Thompson and Cosgrove, in press), this model for the evolution of the breccia does not explain the organised distribution of clasts within the breccia. Therefore further non-brecciation processes must be included to account for this aspect of the breccia.

BRECCIA DESCRIPTION

The breccia consists of a complex series of layers and channels (photo 1, fig. 4), which contain matrix supported clasts of the surrounding shales and re-brecciated breccia. The clasts tend to be rounded to subrounded if they are shale and extremely well rounded if they are re-brecciated breccia. Clast:matrix ratios are extremely variable from approximately 70-80% clasts to 70-80% matrix (photo 2), though it is more common to have a higher percentage of matrix than clast material. The highest clast:matrix ratios (photo 5) are often associated with intensely slickensided low angle slip planes (top of photo 5). This results from a combination of a great decrease in clast size with a great increase in spacing between clasts. The layering ranges in style from extremely clearly defined bands, usually delineated by thin dark bands, to bands with no boundaries at all in the matrix. These layers are defined purely on a clast association (photo 3/4). Within each layer there may be clast distributions which are:

a. of uniform size and distribution.
b. of variable size and uniform distribution (photo 3).
c. of variable size and graded distribution (photo 1/4).

There are two classes of graded distribution, fining downwards and bi-directional, fining outwards towards the boundaries of the layer. Visual examination of the breccia suggests that there is a preferred orientation of the clasts with a high aspect ratio (photo 4).
One of the most unusual features of the breccia are the growth textures in the matrix (photo 6). The quartz crystals are euhehedral and typically grow perpendicular to the surface of nucleation, these surfaces being vein walls, layer boundaries and clast surfaces. There is a layer of cryptocrystalline quartz at the country rock/matrix interface, which may have a layer of anhedral, fine grained quartz overgrowth. The euhehedral crystals grow from this layer. Where crystal growth nucleates on a clast surface, a corona of radial euhehedral crystals is produced, rooted in this layer of cryptocrystalline or anhedral quartz. Even on planar surfaces clusters of crystals are often radiate in form, each individual crystal growing wider with increasing distance from the nucleation surface as some crystals develop at the expense of others. Examination of these coronas reveals multiphase development, with as many as four phases of growth visible. Often each layer of euhehedral growth has an associated anhedral layer, coating the previous euhehedral layer, suggesting that the processes which modified the breccia encompassed a two phase crystallization process, rather than a single initial anhedral phase, followed by a repetitive process of euhehedral crystallisation. Despite extensive mineralization, little of the quartz growth takes the form of pore space infills, being concentrated into surface-normal growth. As a result the breccia maintains a high degree of permeability.

FORMATION/EVOLUTION

The original breccia is currently envisaged to have formed as a result of extreme dilation, causing implosion of the shales. This generated a chaotically arranged assembly of irregular clast (photo 7). These clasts were already extensively hydrofractured and veined due to fluid accumulation and overpressuring during earlier thrusting.

In the proposed model, the present form of the breccia is almost entirely due to the effects of reworking and alteration. The model utilises repetitive seismic events and the resultant fluid intake to activate sorting processes within the clast assemblage (figure 4). The vibration from a failure event could, in itself, impart a degree of ordering to a loosely consolidated breccia (Barker and Mehta, 1993). In addition, the stress release and pressure drop associated with a failure event draws fluids into the fault zone from the surrounding rocks (Sibson 1990). Because the breccia is unconsolidated it would be possible for it to have become fluidised by a pulse of high velocity fluid. This process would mobilise the clasts, making it possible for them to be organised by velocity variations within the fluid. Variations in clast size and shape and the void:clast ratio would define flow channels, in the form of complex and highly changeable layers/cells. Each layer would be sorted internally by the interacting processes of vibration and fluidization. Fluidization of layers is a powerful mechanism to help explain the present form of the breccia for two reasons. Firstly, it is capable of producing a graded clast distribution by flow, a plausible mechanism for forming bi-directional fining out distributions. Secondly, it potentially creates a suitable environment for the formation of a silica gel, (Davidson and Harrison, 1971, Herrington and Wilkinson, 1993, Stel and Lankreyer, 1993) derived from the pore fluids mobilised by seismic activity. Gellification is a rapid process and would provide a crystallizing medium of sufficient viscosity to create the matrix supported texture exhibited by the majority of the breccia, without disordering the clasts. The variations in fluid intake and clast assemblage which produce localised fluid flow pathways reinforce layer development and enhance the channel-like structure of some layers. Repeated fluid intake events will produce repeated reworking events which will gradually mineralise the entire breccia body. This will result in the loss of its highly porous loosely consolidated structure and it will start responding to stress as a rigid body. The rapidity of this consolidation process will be controlled by the fluid supply and the intensity of seismic activity, as well as the original clast size, shape and distribution.
Once the breccia begins to deform as a rigid body, the low angle slip planes (fig. 4), become active. These channelled the flow of later fluid pulses. The zones adjacent to these slip planes show very high clast:matrix ratios and a very widely spaced clast distribution (photo 5). This texture suggests an original distribution similar to the rest of the breccia which has been subjected to intense clast alteration and mineralization, centred on the slip plane. The slickensides on the planes correspond precisely to those related to the late extension episode and these, together with similar faults crosscutting the breccia at a much smaller scale (figure 3), demonstrate that the breccia was acting as a fluid sink, undergoing modification and mineralization throughout the Variscan orogeny.

DISCUSSION

The current work on the Rusey Breccia shows that it is possible, given an original episode of great dilation, to produce the structures and textures observed without requiring unique events or processes. The mechanisms and processes described in the previous section, episodic failure and fluid intake, should operate in most active fault systems.

The processes of fluidization are well documented in the sedimentological literature (e.g. Lowe, 1976. Nichols, 1995), and the ordering of particles by vibration and fluidised flow regimes are discussed in engineering literature (Davidson and Harrison, 1971, Barker and Mehta, 1993). These processes require an unconsolidated substrate to be viable and work most effectively in material with large amounts of voidage.

For these processes to be relevant to the modification of fault breccias, the breccia would have to be highly unconsolidated, requiring a significant volume increase between the unbrecciated and brecciated states. There is considerable evidence for high levels of dilation in and around the Rusey Fault, such as patterns of brittle failure and mineralization resulting from a uniform expansion of the rock volume. The periphery of the breccia band shows numerous examples of thick (3-8cm), randomly oriented veins containing wall-normal crystal growth and matrix supported clasts. The clasts are angular and can often be traced to their pre-brecciation location. They appear to be typical products of hydrofracture, albeit with large amounts of space created and mineralised. There are common examples of ladder veins, where brittle failure and extension has occurred both vertically and horizontally. The matrix supported structure of the Rusey breccia itself requires either a large degree of expansion or considerable removal of material from the system. The evidence for both a tensional stress regime and high pore fluid pressures makes the former a plausible model. The breccia is still extremely porous, indeed vuggy, showing episodic mineralization, implying that there was originally far more void space than at present. It is therefore justifiable to propose that the original breccia would be responsive to modification by the processes described above.

In the case of the Rusey Breccia, the unconsolidated nature of the initial breccia serves to greatly enhance the effect of the modifying processes. The Rusey Breccia, therefore, provides an insight into the effects of extensive fluid flow which couldn't be gained from a more consolidated breccia. The mobilisation of the breccia clasts by metamorphic and meteoric fluids was highly localised into layers and channels. This has important implications for the distribution of fluids and the evolution of deformation associated with fault zone breccias.

CONCLUSIONS

- The formation and evolution of the Rusey Breccia can be confidently correlated to the larger scale tectonics of the Variscan orogeny (Thompson and Cosgrove, in press).
• The breccia was formed under unusually dilatant conditions, generating a zone of high porosity and permeability.
• Intake of fluids into the brecciated zone initiated fluid flow processes, thereby modifying the original breccia distribution.
• The combination of high porosity/permeability and high fluid intake resulted in the formation of a matrix supported system, permitting unusual growth textures in the matrix.

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Figure 1. The South-west peninsula and Rusey

- Lizard complex
- granite
- thrust fault
- normal fault
Figure 2. Structural relationships within the breccia
<table>
<thead>
<tr>
<th>FLUID TYPE</th>
<th>SOURCE</th>
<th>LOCATION</th>
<th>STRESS STATE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>metamorphic/basinal brines</td>
<td>veins in clasts</td>
<td>THRUSTING DURING CONVERGENCE</td>
</tr>
<tr>
<td>2</td>
<td>mixed metamorphic and meteoric water</td>
<td>matrix</td>
<td>STRIKE-SLIP AS CONVERGENCE CEASES</td>
</tr>
<tr>
<td>3A</td>
<td>meteoric with some metamorphic</td>
<td>euhedral crystals growing radially around the clasts</td>
<td>POST-OROCgenic COLLAPSE</td>
</tr>
<tr>
<td>3B</td>
<td>meteoric</td>
<td>low angle tensional fractures</td>
<td></td>
</tr>
<tr>
<td>3C</td>
<td>meteoric</td>
<td>high angle shear fractures</td>
<td></td>
</tr>
</tbody>
</table>
Figure 3. Structural map of the Rusey Headland
Figure 4. Evolution of the Rusey Breccia
Plate 1. Layers within the breccia.

Plate 2. Variations in clast:matrix ratio within the breccia.
Plate 3. Ungraded clast layers within the breccia
Plate 4. Graded clast layers within the breccia
Plate 5. Alteration zone adjacent to low angle slip plane
Plate 6. Growth textures in the matrix
Plate 7. Hydrofractured breccia
Palaeo and Present-day Fluid Flow through Eocene Clay Layers in Flanders

Hydrogeological and Hydrogeochemical Evidence for the Present-day Existence of Preferential Pathways in the Bartonian Clay

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Abstract

The semi-confined Ledo-Paniselian (Eocene) aquifer in Flanders is recharged in the areas with a higher topography, where it is covered by the Bartonian clay [1]. Recharge is thus occurring by downward groundwater flow through the Bartonian clay. This is demonstrated by piezometric levels. Flow modeling in the recharge area of Ursel, where many piezometers provide an excellent knowledge of the hydraulic heads, has indicated a vertical hydraulic conductivity for the Bartonian clay of $10^7$ m/s. However, laboratory measurements often provide values which are at least one order of magnitude lower. This discrepancy can be ascribed to the presence of preferential pathways in the clay, through which the flow is preferentially taking place.

The considered Tertiary sediments have all been deposited in a marine environment. At the end of the Tertiary, the sea regressed from the area and the present-day topography developed due to fluvial erosion. Groundwater movement was induced, by recharge of precipitation in the higher regions. Calcite dissolved in the infiltrating water, and freshening of the sediments took place, consisting both in dilution of the marine pore solution and in cation exchange, the latter mainly in the Bartonian clay in the recharge areas [2]. The early formed groundwaters were pushed to the north, in the direction of groundwater movement in the aquifer, as more precipitation was infiltrating in the recharge areas. In the aquifer, in an upstream direction, progressively more freshened watertypes are found. The chromatographic sequence of cation exchange is expressed by the subsequent surplus of Na$,^+$, followed by K$^+$ and finally Mg$^{2+}$, resulting in NaHCO$_3$ and MgHCO$_3$ watertypes. The groundwater leaking out of the clay and entering the aquifer nowadays in the recharge area, contains only the Ca$^{2+}$ cation in appreciable quantities.

A cored boring in the recharge area at Ursel has provided the opportunity for determining CEC and exchangeable cations, both of the clay and of the underlying aquifer [3]. The Bartonian clay at Ursel shows a very clear depletion in adsorbed Na$,^+$, and to a lesser extent also in K$^+$, when compared to a boring out of the recharge area. Adsorbed Ca$^{2+}$ is higher at Ursel. However, the analyzed samples of the Bartonian clay at Ursel do not at all show a depletion in adsorbed Mg$^{2+}$. This is a surprising result,
as the Ledo-Paniselian aquifer in this recharge area is containing CaHCO₃ water, indicating that the flushing of the overlying clay layer has practically been accomplished [4].

The geochemical/mixing cell model PHREEQM [5] has been used to simulate the freshening of the Bartonian clay [4] and the subsequent recharge to the underlying aquifer [6]. After the calculated flushing of the Bartonian clay by around 100 pore volumes, the distribution of the exchangeable cations in the clay is similar to the one found in the boring at Ursel. At that stage the pore water solution which is leaving the Bartonian contains Ca²⁺ and Mg²⁺ as main cations and no significant Na⁺ content. This water is strongly different from the one found at present in the considered recharge area of the Ledo-Paniselian aquifer, in which Mg²⁺ is almost absent. When the different solutions leaking out of the clay till this stage are used to flush the aquifer, it is not possible to model the observed distribution of the groundwater quality in the Ledo-Paniselian. The clay must be flushed near 400 times to obtain the present situation in the aquifer. This confirms the existence of preferential paths in the Bartonian clay, where the flow is faster. The flow line in the analyzed boring must be part of a slower pathway and the freshening here is still in progress.

INTRODUCTION

The survey area comprises the northern part of East- and West-Flanders in Belgium (fig. 1). The Tertiary layers consist of alternating sand and clay. They are gently dipping to the NNE. Recharge of the sandy Ledo-Paniselian aquifer is occurring in the highest regions in the south of the survey area, by infiltration through the overlying Bartonian clay.

MODELING OF GROUNDWATER FLOW

The hydraulic conductivities of the sandy sediments have reliably been obtained from pumping tests. The values for the clayey sediments have posed more problems. Laboratory tests indicate values ranging around 10-5 m/d or 10-10 m/s, or even lower [1].

In the westerly recharge area around Ursel, the hydraulic head above and below the Bartonian clay is accurately known, by the presence of 56 piezometers. The observed hydraulic head below it could not be simulated by assuming a vertical hydraulic conductivity of 10-10 m/s for the Bartonian clay. Calculated heads were much too low. The local thickness of the clay being known, its hydraulic conductivity could be deduced by calibrating the model on the observed hydraulic head distribution. An excellent agreement between observed and calculated heads has been obtained with a value of 10-9 m/s [1].

The discrepancy between hydraulic conductivity values obtained from laboratory tests on local samples and the values deduced from the approach, based on the regional hydraulic head distribution, has been ascribed to the presence of preferential pathways in the clay, through which the flow is preferentially taking place.

The calculated groundwater flow in natural conditions confirms that the aquifer is mainly recharged by infiltration through the Bartonian clay. From the recharge areas on, groundwater generally flows towards the north to north-west. Outside the recharge areas, a very slow upward flow occurs, along which groundwater is leaving the aquifer by flowing out through the Bartonian clay. This vertical outflow is gradually reducing the velocity within the aquifer, in the flow direction [7].
Figure 1. Geology of the survey area and groundwater flow.
HYDROGEOCHEMICAL EVOLUTION

The considered Tertiary sediments have all been deposited in a marine environment. At the end of the Tertiary, the sea regressed from the area and the present-day topography developed due to fluvial erosion. Groundwater movement was induced, by recharge of precipitation in the higher regions. Calcite dissolved in the infiltrating water, and freshening of the sediments took place, consisting both in dilution of the marine pore solution and in cation exchange, the latter mainly in the Bartonian clay in the recharge areas [2]. The early formed groundwaters were pushed to the north, in the direction of groundwater movement in the aquifer, as more precipitation was infiltrating in the recharge areas.

The groundwater quality distribution in the Ledo-Paniselian aquifer is resulting from this evolution, whereby the marine conditions are being expelled by the infiltrating fresh water (fig. 2). In the westerly recharge area around Ursel, the groundwater type is CaHCO$_3$. Downward the flow (to the NNE), the MgHCO$_3$ type appears. More towards the north, the NaHCO$_3$ type is found, and still further northward the NaCl type. In the meantime, salinity increases towards the north, from fresh (F), fresh-brackish (Fb), brackish (B) to brackish-salt (Bs). Thus, in an upstream direction, progressively more freshened watery pes are found. The chromatographic sequence of cation exchange is expressed by the subsequent surplus of Na$^+$, followed by K$^+$ and finally Mg$^{2+}$, resulting in the NaHCO$_3$ and MgHCO$_3$ watertypes. The groundwater leaking out of the clay and entering the aquifer nowadays in the recharge area, contains only the Ca$^{2+}$ cation in appreciable quantities.

The sequence of groundwater types observed in the Ledo-Paniselian aquifer is in excellent agreement with the pattern of natural groundwater flow (fig. 1).

A cored boring in the recharge area at Ursel has provided the opportunity for determining CEC and exchangeable cations, both of the clay and of the underlying aquifer [3]. The determination of the CEC and the extraction of the exchangeable cations were performed by means of NH$_4^+$-acetate at pH 7. As the salts soluble in water also dissolve in NH$_4^+$-acetate, they were determined separately, and the result was subtracted from the one in NH$_4^+$-acetate. CEC for the Bartonian clay at Ursel ranges around 27 meq/100g and for the Ledo-Paniselian aquifer, CEC is 5.5 meq/100g on the average (fig. 3). The exchangeable cations in the Bartonian clay at Ursel were determined to be (fig. 4): CaX$_3$ = 12 meq/100g, ranging between 5-18 meq/100g; MgX$_3$ = 13 meq/100g, ranging between 7-19 meq/100g; and less than 3 and 0.2 meq/100g for KX and NaX respectively [3].

The Bartonian clay at Ursel shows a very clear depletion in adsorbed Na+, and to a lesser extent also in K$, when compared to a boring out of the recharge area, at Assenede (fig.4). Adsorbed Ca$^{2+}$ is higher at Ursel. The adsorbed cations confirm that the Bartonian clay has been leached out to a large extent in the recharge area. Out of the recharge area, the marine influence still persists, expressed by a high concentration of NaX.

However, the analyzed samples of the Bartonian clay at Ursel do not at all show a depletion in adsorbed Mg$^{2+}$. This is a surprising result, as the Ledo-Paniselian aquifer in this recharge area is containing CaHCO$_3$ water, indicating that the flushing of the overlying clay layer has practically been accomplished [4].
HYDROGEOCHEMICAL MODELING

The geochemical/mixing cell model PHREEQM [5] has been used to simulate the freshening of the Bartonian clay [4] and the subsequent recharge to the underlying aquifer [6]. A flow line has been selected, starting from the top of the Bartonian clay in the westerly recharge area at Ursel, and directed towards the NNE within the aquifer. Model results have been compared with groundwater quality observations of the aquifer, taken from the rectangular area indicated in figure 2. The modeling has allowed to show that the present groundwater quality distribution in the Leda-Paniselian aquifer mainly corresponds to cation exchange processes in the overlying Bartonian clay and, to a smaller extent, also within the aquifer. Other effects that have been considered in the model as well, are related with the oxidizing conditions prevailing in the clay, responsible for the observed presence of gypsum, and the reducing conditions in the aquifer, causing sulfate reduction. Calcite precipitation/dissolution processes have also been considered.

Groundwater flow conditions in the Bartonian clay are largely differing from those in the Leda-Paniselian aquifer, with respect to differences in flow velocity and length of flow path (comparing the thickness of the clay, in the order of tens of meters, to the length of the flow path within the aquifer, which is tens of kilometers long). Therefore, the modeling was divided in two parts. First, the freshening of the clay was simulated in a vertical column; subsequently, the different watertypes
leaking out of the Bartonian clay were flushed through a horizontal column of the Ledo-Paniselian aquifer, simulating the subhorizontal flow in it.

An overview of the model boundaries is given in figure 5. For both clay and aquifer, the initial pore water quality was sea water, equilibrated with calcite and 0.75 mmol/kg H$_2$O of NH$_4^+$ production. This solution was also equilibrated with the exchange complex in the clay and in the sandy aquifer, respectively. For the recharge water, pure water equilibrated with calcite and a $P_{CO_2}$ of 10$^{2.0}$ atm, was used. The flow velocities were deduced from the flow model [7]. The cation exchange parameters were computed from data in [1] ([8]).

![Figure 3 - CEC in sediments of cored borings at Ursel and Assenede.](image-url)
Figure 4a: Assorted cations (Na⁺ and K⁺) in sediments of cores bored at Ursel and Assenede.
Figure 4b - Adsorbed cations (Mg$^{2+}$ and Ca$^{2+}$) in sediments of cored bores at Ursel and Assenede.
The evolution of the distribution of exchangeable cations adsorbed to the Bartonian clay, and of the composition of the porewater leaking out of it during the flushing, are shown in figure 6. After the clay has been flushed 100 times, the distribution on the clay’s adsorption complex is similar to the one found in the boring at Ursel. At that moment, the pore water solution leaving the clay contains Mg$^{2+}$ and Ca$^{2+}$ as main cations, with no significant Na$^+$ content. This water is strongly different from the one found at present in the Ledo-Paniselian aquifer in the recharge area of Ursel, in which Mg$^{2+}$ is almost absent. The clay must be flushed near 400 times to obtain this observed watertype. This confirms the existence of preferential pathways in the Bartonian clay, through which flow is faster. The flow line in the analyzed boring must be part of a slower pathway, where the freshening is still in progress [4].

When the different solutions leaking out from the Bartonian clay, after flushing it 100 times, are used to flush the Ledo-Paniselian aquifer, the modeled groundwater quality distribution in the aquifer does not correspond at all to the observations (fig. 7). The clay must be flushed near 400 times to obtain the present distribution in the aquifer (fig. 8). This is again confirming the previous conclusion. The deviation between model results and observations, which can be noticed in figure 8 for the most northward part of the aquifer (most downstream 5 km), is resulting from the lower velocities in the deeper parts of the aquifer [8], which are not accounted for in the modeling results presented here.

### HYDRAULIC AND HYDROCHEMICAL CONDITIONS FOR MODELING THE FRESHENING OF THE BARTONIAN CLAY AND LEDO-PANISELIAN AQUIFER.

<table>
<thead>
<tr>
<th>Infiltration water</th>
<th>Initial pore water (sea water equilibrated with calcite and 0.75 mM NH$_4^+$ production):</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca$^{2+}$ = 2.1 mM/l</td>
<td>Ca$^{2+}$ = 9.8</td>
</tr>
<tr>
<td>HCO$_3^-$ = 4.17 meq/l</td>
<td>K$^+$ = 10.6</td>
</tr>
<tr>
<td>pH = 7.3156</td>
<td>Cl$^-$ = 568 (in mM/l)</td>
</tr>
<tr>
<td>lgP$_{CO_2}$ = -2.00</td>
<td>SO$_4^{2-}$ = 29.3</td>
</tr>
<tr>
<td>Na$^+$ = 488</td>
<td>NH$_4^+$ = 0.75</td>
</tr>
<tr>
<td>HCO$_3^-$ = 1.6</td>
<td>pH = 7.86</td>
</tr>
</tbody>
</table>

**- BARTONIAN CLAY**

- Column length: 20 m
- Number of cells: 4 (5 m each one)
- Flow velocity: 0.12 m/year
- Time step: 41.7 years
- Dispersivity: 1.5 m
- Diffusion coef.: $10^3$ m$^2$/s
- Cation Exchange Capacity: 27 meq/100 gr.
- Gypsum dissolution: 0.45 mM/cell
- Calcite equilibrium

Exchange parameters (computed from data in Walraevens, 1987).

**- LEDO-PANISELIAN AQUIFER**

- Column length: 20 km
- Number of cells: 20 (1 km each one)
- Flow velocity: 3.36 m/year
- Time step: 299 years
- Dispersivity: 1500 m
- Cation Exchange Capacity: 5.5 meq/100 gr.
- $SO_4^{2-}$ reduction: 1.7 mM/cell in the first 4 cells
- Calcite equilibrium

Exchange parameters [Standard coefficients in PHREEQI).

Figure 5 - Model boundaries for freshening of Bartonian clay and Ledo-Paniselian aquifer.
Figure 6 - Evolution of exchangeable cation distribution and of composition of porewater leaking out of Bartonian clay during flushing.
Figure 7. Comparison of observed concentrations of main cations in Ledo-Paniselian aquifer, to modeled concentrations after flushing Bartonian clay 100 times.

Main Cations Contents in the Ledo-Paniselian Aquifer along the Flow Path, and Modeled Values after the Bartonian Clay Has Been Flushed 100 Times.

- **Ca²⁺**
  - Concentration (mg/l) vs. Distance (km)
  - X model calculation
  - O observation

- **Na⁺**
  - Concentration (mg/l) vs. Distance (km)
  - X model calculation
  - O observation

- **Mg²⁺**
  - Concentration (mg/l) vs. Distance (km)
  - X model calculation
  - O observation

- **K⁺**
  - Concentration (mg/l) vs. Distance (km)
  - X model calculation
  - O observation
Figure 8 - Comparison of observed concentrations of main cations in Ldeo-Paniselian aquifer to modeled concentrations after flushing Bartonian clay 400 times.

Main Cations Contents in the Ldeo-Paniselian Aquifer along the Flow Path, and Modeled Values after the Bartonian Clay Has Been Flushed 400 Times.

- Ca²⁺
- Na⁺
- Mg²⁺
- K⁺

* model calculation  o observation
Palaeo and Present-day Fluid Flow through Eocene Clay Layers in Flanders

Fracturation and Intraformational Faulting of the Ieper Clay: Evidence for a Major Palaeo-flow Event

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Abstract

The microfracturation of the Eocene London or Ieper Clay and of the Oligocene Boom Clay has been well known to soil mechanics engineers for more than a century. Some authors [9] have related the microfracturing of the London Clay to local basement-induced tectonics. This hypothesis seems to be invalidated because microfracturing in the equivalent Ieper Clay is also observed in areas in Belgium where no basement-induced tectonic deformations are known.

In addition, high-resolution reflection seismic investigations have revealed large-scale intraformational fault patterns in the London or Ieper Clay over their full extent in the Southern North Sea [10] and even over a large area of the basin axis in the Central and Northern North Sea [11]. Such patterns involve faults with a throw of several metres, sometimes up to 10 m, which however are confined to the clay layer and fade away towards its base and top boundary.

Fault patterns have also been observed on quarry profiles in South Flanders, and are apparently similar to those observed on seismograms. An outstanding example is found in the Koekelberg quarry in Marke. The fault surfaces are generally characterized by a well-developed fault mirror with striae and a black fault gauge. A striking feature has been the discovery of microfossils within such gauge, originating from clay horizons about 60 m lower.

These observations have led to a model, involving a build-up of undercompaction in the early burial history of the clays, and its subsequent relaxation through hydraulic fracturing, at fairly shallow depths [12]. The sealing of a pressure compartment would initially be established by preferential fluid drainage and compaction of units adjacent to more permeable strata. The development of a density inversion due to undercompaction resulted in a Rayleigh-Taylor instability, for which clues have been found in the Southern North Sea. This model has been endorsed by the Imperial College group, which investigated three-dimensional high-resolution seismic data in the Central and Northern North Sea [11].

This field evidence of an early major hydraulic fracturation and dewatering event, which left its scars under the shape of a general network of faults and intense microfracturing, deserves attention in any evaluation of the seal capacity of the Ieper Clay under renewed hydraulic and/or thermal stresses.
Land observations

The microfracturation of the Eocene London or Ieper Clay and of the Oligocene Boom clay has been well known to soil mechanics engineers for more than a century. Some authors [5] have related the microfracturing of the London Clay to local basement-induced tectonics. This hypothesis seems to be invalidated, first of all because microfracturing in the equivalent Ieper Clay is also observed in areas in Belgium where no basement-induced tectonic deformations are known.

In various clay quarries in South Belgium (in Marke, Lauwe, Zonnebeke, etc.), microfracturation is associated with large-scale faulting (fig. 9). Throws are up to several metres, and dips ranging from 45 to 80 degrees. The fault surfaces in Marke (Koekelberg quarry) are generally characterized by a well-developed fault mirror with striae and a black fault gauge [15], from some fractions of a millimetre up to 15 mm thick. A striking feature has been the discovery of microfossils within such gauge, originating from clay horizons about 80 m lower, arguing for a major fluid expulsion event.

Figure 9 - Faulting in Ieper clay, Koekelberg clay pit, Marke, Belgium [15].
Some observations of faults in Marke reveal also a secondary, dense set of faults, with relative displacements in the centimetre to decimetre scale, often in strike-slip mode. They are characterized by the absence of striae and gauge, and are probably to be considered as decompression features, related to the uplift and erosion of the Paleogene sequence. At the top of the quarries, faults often seem to have been reactivated by periglacial phenomena in Quaternary times. This periglacial overprint is easy to differentiate from the primary deformations, but demonstrates how such inherited features can be reactivated locally under new stress conditions.

**Marine observations**

The major clue towards the general, basin-wide evidence and significance of such fracturation in Cenozoic clays in the North Sea Basin has however come from high-resolution marine reflection seismic investigations. A systematic mapping of the Southern North Sea by RCMG has revealed general, large-scale intraformational fault patterns in the London or Ieper Clay [10], over its whole stratigraphic thickness (140 m) but with a vertical zonation in deformation style.

The lower interval of the Ieper Clay, up to about 25 metres above the undisturbed basal reflector of the clay, is generally characterized by a dense pattern of block faulting, with tilted and arched blocks and apparently randomly dipping fault planes. The average throw is about two metres.

In a second interval, from about 25 metres to up to at least 70 metres above the clay base, one may locally observe a well-preserved wavy deformation pattern, consisting of a festoon-like alternation of broad, rounded synclines and narrow, cuspatate anticlines. Such anticlines seem to develop locally into diapiric structures. Again, one diapiric feature pierces the base of a Quaternary palaeovalley, arguing for a surficial periglacial reactivation of primary deformations. The average wavelength of the festoons amounts to a few hundreds of metres, and the amplitude ranges from 2 to 10 metres.

The most general deformation style observed however is a pattern of faulted and tilted blocks, sometimes with a dominant tilt direction. Approximately where the Ieper Clay grades into the overlying Ypresian sands, about 140 m above the base, another peculiar deformation pattern is observed, involving faulted blocks without noticeable block movement but with inverse drag features along the fault-planes.

The deformations observed on seismograms in the roof zone of the Ypresian Clay progressively fade away in the overlying sands.

Recently, Cartwright [11, 14, 16] has described large-scale intraformational faulting patterns within a 200 to 1000 m thick sequence of Palaeocene to early Miocene shales in the Central and Northern North Sea. Faults with throws ranging from 10 to 100 m could be imaged by industrial 3D surveys. The average fault spacing is 200-500 metres, and the average dimensions of the faults are 500 – 1000 metres in map trace length. The dips exhibited by the faults range from 30 degrees to subvertical. The strike orientation of the faults do not exhibit any strong preferred direction. Just like on the Belgian continental shelf, in particular in the upper interval of the Ieper Clay, on cross sections oriented down the palaeoslope, the majority of faults have hanging walls that face in an up-slope direction.

The most important structural observation derived from the interpretation of the 3D data is that the extensional faults are arranged in polygonal networks (fig. 10).
Figure 10 - Surface distribution of faults on a detailed study of the Ieper/London clay north of North Hinder Bank (Southern North Sea) [15] and block diagram showing the 3-D fault geometry in Early Cenozoic mudstones in the Central North Sea [16].

Genetic model

The observations in the Ieper clay have led to a model, involving a build-up of undercompaction in the early burial history of the clays, and its subsequent relaxation through hydraulic fracturing and de-watering, at fairly shallow depths (fig. 11) [12].

The Ypresian clays were deposited as high-porosity, water-logged muds. As sedimentation progressed and the thickness of the clay deposits increased, pore space gradually decreased through the expulsion of pore water. Pore water drainage was probably greatest near the base, due to the maximal overburden weight and the proximity of a permeable basal bed. The fast drainage of the basal beds of the clay developed a permeability barrier, soon impeding further basal drainage.

When clay sedimentation gave way to the deposition of sands, one might imagine that a similar sealing process occurred at the top of the Ieper Clay sequence. Rapid drainage through the permeable sand cover could induce compaction from the top down, in addition to the compaction from the bottom up.
As a result, the clay may have sealed itself, with two related consequences:

(1) as water has a low compressibility, the sealed part of the clay will have remained undercompacted for a time, with a lower density than its overburden of compacted clays and sands; this density inversion is gravitationally unstable;

(2) as soon as drainage was impeded in some part of the clay body, continued sedimentation implied that the locked pore water became overpressured; the resulting decrease in effective normal stress acting on the interparticle contacts decreased the shear strength of the sediment.

Both processes together may be regarded as principal agents in the development of clay tectonic deformations such as those observed in the Ieper Clay, and especially of the clay waves. The gravitational instability probably acted as the motor, which drove the sediment flow, while the overpressured pore water acted as a lubricant, decreasing the shear resistances at the grain contacts.

Figure 11 - Genetic model of the deformation of the Ieper/ London clay [12].
The development of a density inversion due to undercompaction resulted in a Rayleigh-Taylor instability, for which clues have been found in the Southern North Sea (the "frozen" festoon-like undulating deformations).

Overpressure in shallow sediments is however intrinsically transitory. Hydrostatic conditions are restored either by slow seepage or by fracturing of the permeability barriers. The fracturing of the compacted (and hence more brittle) clay beds above the main undercompacted horizon could have taken place both by progressive hydrofracturing and by local fault development, induced by the buoyant force of the upwelling clay crests. As pore pressure relaxed, the upper, brittle horizons underwent faulting and tilting while progressively sagging into the initially undercompacted horizons.

The basic principle of this model has been endorsed by the Imperial College group [11], and further developed for the more comprehensive sequences of the Central North Sea Basin into a model for episodic basin-wide fluid expulsion from geopressed shale sequences. The fault polygons observed in three dimensional seismic surveys could have been produced by the spatial arrangement of shale flow structures triggered by the density inversion into a pattern of polygonal “dome and sink” features analogous to those produced experimentally [13].

**Implications**

This general field evidence of early major hydraulic fracturation and dewatering events, possibly of episodic nature in comprehensive sequences, deserves attention in various fields of fundamental and applied significance.

1. In the appraisal of the dynamics of oil source rocks and reservoirs, this evidence of very early hydrofracturation events of shales, at potentially very shallow depths (a possibility which had formerly been virtually completely overlooked by the oil industry), has implications on the timing of the primary migration of hydrocarbons from source rock to reservoirs.

2. Though the faults seem to fade out in overlying layers, their reactivation in subsequent dewatering events at different levels may contribute to the partitioning of intercalated potential reservoirs.

3. The basin-wide collapse of the overpressured compartments can have resulted in a geologically rapid flux of enormous volumes of pore fluids, with possible palaeo-environmental implications. The associated heat flow pulses can play a role in basin evolution.

4. Single-pulse or episodic dewatering can play a role in the diagenesis of overlying horizons, affecting their permeability. Episodic fluid pulses have already been advanced for explaining the petrogenesis of Pb-Zn deposits.

5. Any evaluation of the seal capacity of the Ieper clay under renewed hydraulic and/or thermal stresses (in case of radioactive waste storage) cannot ignore the potential role of these fracture networks.
Conclusion

The above observations of inherited and present-day potential fluid pathways in Eocene clays in Flanders deserve attention with regard to the potential sealing capacity of such clays. The extensive data base already collected and the easy access to quarries, boreholes and extensive outcrops at the sea bed, where these clays can be investigated with continuous profiling techniques, may turn these units into a readily accessible field laboratory.

Hydrogeological and hydrochemical evidence has been deduced for the present-day existence of preferential pathways in the Bartonian clay. A difference in hydraulic conductivity values for this clay layer of at least one order of magnitude, has been obtained between laboratory tests on local samples and a model calibration approach, based on the known regional hydraulic head distribution. Preferential pathways of flow in the clay can account for this difference. Also the apparent incompatibility between the observed adsorbed cations in the Bartonian clay in the recharge area of the Ledo-Paniselian aquifer, and the composition of the groundwater in this aquifer, can be ascribed to the presence of preferential pathways in the clay, through which the flow is faster. The hydrogeological and hydrogeochemical investigation carried out for the Bartonian clay will be conducted for the Ieper Clay, in order to establish whether preferential pathways are indicated here as well. On the other hand, the hydraulic conductivity along observed faults and fractures should be studied.

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An evaluation of the occurrence of fluid flow through faults and fractures is of primary importance for the performance assessment of radioactive waste repositories located in argillaceous settings.

In order to provide the national waste management organisations and the scientific community at large with insights into the driving processes and the occurrence of fluid flow through faults and fractures in argillaceous formations, the NEA and the EC jointly organised a workshop on this topic (Berne, Switzerland, 10-12 June 1996).

This publication includes the papers presented orally or as posters at the workshop, and is introduced by a synthesis of the topics addressed and the conclusions reached.