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SEMINAR ON FISSION GAS BEHAVIOUR IN WATER REACTOR FUELS - EXECUTIVE SUMMARY

Cadarache, France
26-29 September 2000

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Seminar on Fission Gas Behaviour in Water Reactor Fuels

Cadarache, 26-29 September 2000

Executive Summary

Introduction

The need for further improving the understanding of basic phenomena underlying nuclear fuel behaviour has been recognised both by fuel vendors, experts in fuel research in the different laboratories and committees and working groups co-ordinating international activities. The OECD/NEA Nuclear Science Committee has established an Experts Group addressing this issue. This has led to establishing an International Fuel Performance Experiments Database (IFPE) that should help model evaluation and validation. Many years ago the IAEA established an International Working Group on Fuel Performance and Technology (IWGFPT) that led to the FUMEX-1 (Fuel Modelling Exercise) which has had an important impact on code improvements. Both international organisations, with the support of national organisations, co-operate in establishing and maintaining the Database and to build confidence in the predictive power of the models through international comparison exercises. But above all the different parties have agreed that seminars focussed on specific phenomena would be beneficial to exchange current knowledge, identify outstanding problems and agree on common action that would lead to improved understanding of the phenomena. A series of such seminars has been initiated by the Commissariat à L'Energie Atomique (CEA), Electricité de France (EdF), Framatome and Cogéma under the aegis of the OECD/NEA and the IAEA.

This was the second in a series of three seminars that started with the seminar on « Thermal Performance in Light Water (High Burnup) Fuels» held at Cadarache, France, from 3 to 6 March, 1998. The series will be completed with a meeting devoted to « Pellet-Clad Mechanical Interaction » in 2002.

The aim of this second 'rendez-vous' was to draw up a comprehensive picture of our current understanding of fission gas behaviour and its impact on the fuel rod, under the widest possible conditions. Fission gas behaviour is a leading parameter to the overall fuel rod performance at high burn-up. A sharp increase in fuel temperature due to 'thermal feedback', cladding loading due to high temperature fuel swelling or an end-of-life fuel rod pressure increase are just some of the many possible consequences that may be ascribed to the behaviour of inert gases. For these reasons, the subject has been dealt with since the first steps of the nuclear industry and many basic data have become available. However, there are still areas under investigation and others where further research is required.

The need to understand and to anticipate the way fission gases behave still justifies the implementation of increasingly sophisticated experimental techniques (both in and out of pile). Their results, particularly over the past ten years, have called for more mechanistic modelling to confirm interpretations, specify and estimate physical interactions and extrapolate calculations to different operating conditions. It should be noted that in recent years the implementation of complex numerical schemes or atomistic simulations is being improved due to extended computer capacity.

The objective of this Seminar was, precisely, to emphasise these more recent developments, from both the experimental and modelling points of view

Subject areas

There are two main subject areas. The first one relates to basic properties of inert gases in oxide matrices, such as fission gas volume or intra-granular diffusion coefficients, radiation-enhanced diffusion, etc. The second one pertains to the more macroscopic consequences of the presence of fission gases such as release and swelling. An important aim of the seminar is to contribute to bridging the gap between both areas.

Scope

High burn-up behaviour, MOX and other advanced fuel concepts were addressed within the scope of the meeting. The broadest range of operating conditions (i.e. normal, off-normal and accidental), and the more recently considered long-term storage conditions, were considered as relevant. It is also worth noting that although fission gases figure prominently on the Agenda, data or results pertaining to the behaviour of helium are also treated.

The areas covered by the seminar included:

1. Basic properties

- Diffusion coefficients:
  - Volume and apparent diffusion coefficients
  - Intra-granular diffusion
  - Radiation enhanced/thermal diffusion
- Properties of gas inside bubbles:
  - Bubble-fission fragment interaction
  - Inter and intra-granular resolution and trapping effects
  - Equations of state

2. In-pile fission gas behaviour and impact

- Inter and intra-granular behaviour
- Bubble growth, migration, and pinning
- Fission gas release and swelling under normal, off-normal and accidental operating conditions
- (Thermal and mechanical) impact on fuel rod behaviour
- High burn-up effects, enhanced release at high burn-up, bubble size and concentration with respect to microstructure changes
- Impact of fuel microstructure (UO_2, MOX and advanced fuels)

Experimental results and their interpretation covered :

- In-pile monitoring : pressure transducer, on-line gas flow measurements, others
- Post Irradiation Examinations
  - Any original measurement of gas precipitated in bubbles
  - Experimental estimation of release in the RIM area
  - Inter-granular/intra-granular gas inventory
  - Size distribution of bubbles from quantitative image analysis (SEM, TEM)
  - Determination of gas localisation and gas behaviour through thermal annealing experiments
- Modelling
  - Atomistic level modelling
  - Diffusion models
  - Code validation

The results of the seminar were discussed at a panel during which session chairs presented the results of their specific session. These are presented in the next chapter. The full proceedings will contain, besides this summary, the 38 papers presented at the seminar. The seminar was attended by about 100 participants from 24 countries, representing 46 organisations. Both the programme of the seminar and the list of participants are provided as Annexes.

Panel Summary

Chaired by: J.A. Turnbull

One of the conclusions reached at the final meeting of the IAEA FUMEX code comparison exercise was the need for a series of workshops to discuss data and models on: Fuel Thermal Performance, Fission Gas Release and PCI. The meeting held at CEA Cadarache in September 2000 was the second workshop in this series.

The phenomenon of Fission Gas Release (FGR) from nuclear fuels has been studied for at least 40 years and therefore should have reached a mature state of understanding. It is clear that during this time many questions have been resolved but many are still unclear. The move of commercial reactors to higher and higher discharge burn-up has uncovered features of fuel behaviour previously unseen and hence resulted in new phenomena to be included in the description of the fission gas release processes.

Temperature is the most dominant parameter in controlling fission gas release and the first workshop, which addressed Thermal Performance, discussed the various aspects of this topic such as: gap conductance, fuel thermal conductivity and its degradation with burn-up. It was clear from the Fission Gas Release Workshop that accurate temperature prediction was a pre-requisite to FGR modelling and, thanks to the FUMEX project and the Thermal Performance Workshop, all codes now contained good thermal performance models.

The meeting was divided into four sessions which occupied three days of presentations. The session titles were: *Feedback from Experience, Basic Mechanisms, Analytical Experiments, Industrial Modelling and Software Packages*. Summaries of each session were prepared by session chairpersons and are appended hereto. On the fourth and final morning, there was a session entitled: *Panel Discussion: Conclusions and Perspectives* where each chairperson gave an oral presentation of their session which was discussed among the participants. In view of the 'maturity' of the subject, each chairperson was requested to address the following three questions:

- What do we know, what is new?
- What don't we know, what are the uncertainties and what remains unclear?
- What initiatives should be implemented in the future?

From the first session, *Feedback from Experience,* we know that there is an enhancement of fission gas release at high burn-up, >50 MWd/kg, both in steady state operation and in transient overpower, but there is no accepted phenomenon to explain this. One result of this is that the well-established Halden 1% FGR criterion relating fuel centreline temperature to burn-up overestimates the onset temperature at these levels of burn-up. That is, the temperature for the onset of release at high burn-up is *lower* than predicted by this criterion. It has been observed that high burn-up fuel contains a region of restructured fuel microstructure close to the pellet periphery, the so-called 'rim' structure. It is tempting to use this as a reason for the enhanced release, but a direct correlation cannot be made. Indeed a measure of the ratio of released krypton and xenon isotopes implies that their origin is the hot central regions of the fuel and not the cold plutonium rich restructured region in the pellet rim. The implication of this observation is that the restructured rim serves mainly to increase the thermal resistivity of the fuel, thus increasing fuel temperatures.

A discussion of this dilemma continued into the second session on *Basic Mechanisms*. Investigations have shown that this rim structure consists of a refinement of the original ~10 micron diameter grains into ~0.1 micron diameter grains and the introduction of a micron size population of bubbles which can account for a swelling up to ~10 volume %. In addition, the fission gas concentration in the matrix falls to a low level. The impression of most workers is that the shortfall in fission gas resides in the bubbles and has not been released from the fuel; however the exact distribution of gas remains uncertain. The fission gas release from the rim is not more than 15 to 20%, showing a high retention capacity of such a restructured material[1]. The mechanism of rim formation is not clear: one suggestion is that the build-up of irradiation damage causes the grain refinement and that this is followed by the collection of gas into the porosity. Alternatively, it has been postulated that the porosity is the first to form and that the sub-micron grains are nucleated from the pore surfaces. It remains a matter of debate whether this rim restructuring is beneficial or detrimental.

Several presentations addressed FGR from the rim both in steady-state and transients such as RIA, concentrating on mechanisms and experimental observations. Measurement of krypton and xenon isotopic ratios in released gas was clearly a valuable technique to determine the relative contribution of the rim region to FGR, and application to RIA tests has proved informative. It was concluded that although we know details of this new high burn-up structure at the micron level, it is clear that a goal for the future is to understand it at the nanometer or atomic level and that there was scope for simulation studies using high energy accelerators to reproduce the restructuring without the constraints of time and radioactivity.

A major contribution to the release process is known to be single gas atom diffusion and quite satisfactory models can be constructed using this, along with irradiation resolution from grain boundaries. Many models employ the diffusion coefficient formulated by Turnbull, White and Wise[2] as described in the IAEA Preston Meeting in 1988. Here the authors gave a two term diffusion coefficient with a third low temperature term to be applied close to surfaces and used for short-lived radioactive species. There was much discussion on this topic in the meeting and it transpires that White has an alternative description using only the two high temperature terms with a ‘fractal’ treatment of the surface to account for the difference in kinetics between long- and short-lived species. Unfortunately this has yet to be published in the external literature.

Extensive study of retained caesium and xenon using Electron Probe Micro Analysis (EPMA) carried out at ITU showed that systematically, caesium diffusion was some three times slower than for the rare gases. This is an important result since, in many accident calculations, it is assumed that their diffusivities are comparable; the assumption therefore is reassuringly pessimistic.

In addition we know from electron microscopy that intragranular bubbles are formed within grains and intergranular bubbles and tunnels along grain boundaries. Theoretical studies of krypton atoms in the UO_2 matrix concluded that both krypton and xenon were insoluble thus casting doubt on the existence of ‘thermal resolution’ as a means of moderating absorption of single gas atoms into intragranular bubbles. In the absence of intragranular bubble mobility, such a mechanism is used in several models in order to predict substantial release in transients. It is agreed that observations support only slow or limited random migration of small bubbles, but a ‘directed’ movement towards grain boundaries due to a vacancy concentration gradient was an interesting proposal by Evans. If this mechanism has a significant contribution, the resulting microstructure is quite distinctive and amenable to future testing. Topics requiring further attention were the re-solution of gas atoms from grain boundary bubbles, particularly for large grain fuel where the grain size was greater than the fission fragment range, and the response of grain boundary porosity in fast transients.

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[1] *J. Spino , D. Papaioannou, I. Ray (ITU), D. Baron (EDF),* “Rim formation and fission gas behaviour: some structure remarks”, Session 2 of the proceedings.

[2] *J. A. Turnbull, R. J. White and C. Wise:* “The diffusion coefficient for fission gas atoms in uranium dioxide”, Technical Committee Meeting on Water Reactor Fuel Element Computer Modelling in Steady-State, Transient and Accident Conditions, Preston England, 19-22 Sept. 1988.

It was clear that there was much more information available on these features than currently accessible through the open literature. There was general support for a more open distribution of information and all participants were urged to publish as much information as possible in the interest of a general improvement in models and their application to safe reactor operation. It was noted that although the level of fundamental research in many establishments had been reduced, there were still original and comprehensive works going on in ITU, for example, improving the investigation techniques (micro-hardness tests, lattice parameter measurements). Also, there was a refreshing intake of young engineers and scientists within CEA Cadarache. The participants looked forward to publication of new and original work from these quarters.

As a digression from fission gas, it is clear that helium generation in long term storage of high burn-up fuel can pose a significant problem, in particular for high burn-up MOX. Further data are required on low temperature helium diffusion coefficients.

The session entitled *Analytical Experiments* started with two presentations describing a novel method of determining the disposition of gas between the matrix and the grain boundaries. Such studies complement those performed with X-Ray Fluorescence (XRF) and EPMA and provide vital information for modellers. The techniques still need refining, particularly regarding the effect of small intragranular bubbles on the results, but promise an evaluation of the grain boundary capacity for gas prior to and during interlinkage.

As mentioned previously, Halden have obtained data from high burn-up fuel which suggests that the temperature for the onset of FGR is lower than previously expected. It was clear that further data were required to substantiate this observation. In addition, there was a need to obtain further data on MOX fuel to compare it with UO_2 fuel performance under identical conditions.

Enhanced release at high burn-up can cause the rod internal pressure to exceed the coolant pressure. The effect of this requires attention. So far, experimental data suggests that the gap does not reopen by clad creep-out and positive feedback does not occur. However, the data are sparse and further experiments are necessary.

Both mechanistic and empirical approaches to FGR modelling were presented in the session *Industrial Modelling and Software Packages.* It is clear that empirical modelling can be very valuable, but -is limited in applicability and is essentially only valid within the confines of parameters and irradiation conditions covered in the database on which it is developed. Modellers were also cautioned about employing multiple mechanistic models and expecting to obtain good predictions by using appropriate fitting parameters. There is a case for independent assessment of mechanistic models and their supporting data before consideration for inclusion within fuel performance codes.

The requirement for further data comparing UO_2 and MOX behaviour was re-affirmed in a paper by Struzik of CEA, which showed that there was a significant difference in behaviour in ramp test above 30 MWd/kg. Below 30 MWd/kg the differences in behaviour could be explained in terms of the radial power profile but at higher burn-up FGR and swelling in MOX was greater than expected.

In conclusion, there was unanimous agreement that the Workshop had been a success with an excellent range of presentations which initiated extensive and in-depth discussion. It is clear that the topic of fission gas release modelling was in a mature state with agreement about most of the important mechanisms contributing to the phenomena. High burn-up and MOX behaviour are the current challenges where there is an incomplete understanding, but the meeting produced some good suggestions for the focus of future work. Participants were urged to publish their work in the open literature so that all could benefit in their quest towards the goal of safe operation of commercial reactors.

Summary of Session 1: Feedback from Experience

Chaired by: M. Trotabas (COGEMA) and M. Billaux (Siemens PC)

Five technical papers were presented at this session. They cover fission gas release measurements for a large variety of UO_2, MOX and gadolinia fuels in steady-state and transient conditions up to very high burnups.

In particular, complete post-irradiation examination results have been obtained on PWR fuel irradiated up to 100 MWd/kgU and on BWR and VVER fuels transiently tested at burnup as high as 60 MWd/kgU. These examinations include fractional fission gas release, xenon and cesium radial distributions by EPMA, ceramography interpretation by image analysis, and investigation of the pellet rim region by scanning electron microscopy.

The main results of these measurements is the observation of a significant increase of fission gas release at high burnup. In steady-state irradiation low temperature fission gas release increases from a few percent at 50 MWd/kgU to about 25% at 100 MWd/kgU. Considering the low power level reached at such a high burnup (140 W/cm, 1000°C centreline temperature) it is difficult to explain that result by thermal diffusion only. These high burnup rods also present a well-developed high burnup structure in the rim region, which extends over more than 1 mm at 100 MWd/kgU. In the outer region of the rim, the bubble size reaches 5 µm and the total porosity about 20%.

In ramp conditions, fractional fission gas release of 50% was measured in VVER rods irradiated to 60 MWd/kgU. The ramp terminal level was as low as 340 W/cm. At 50 MWd/kgU the same release was obtained at 440 W/cm. In the latter rod, the temperature was high enough to close the as-fabricated central hole and to form a new hole by columnar grain growth mechanism. The central thermocouples showed temperatures in excess of 1500 °C. The difference of release at 50 and 60 MWd/kgU cannot be explained by the degradation of fuel thermal conductivity only. Similar results have been obtained with NFD BWR fuel irradiated at 61 MWd/kgU.

The conclusion of this session is that a significant enhancement of fission gas release is observed at high burnup in both steady-state and transient conditions. This enhancement can be correlated with the extensive development of a high burnup structure in the rim region. A new mechanism seems to be necessary to explain quantitatively the acceleration of fission gas release above 50 MWd/kgU.

Summary of Session 2: Basic Mechanisms.

Chaired by: R J White (BNFL) & J Rouault (CEA).

Fifteen papers were presented on the basic mechanisms of fission gas release. Two principal areas were discussed in detail. These were the mechanistics of fission gas release and swelling under thermally activated conditions, and the structure and response of the high burnup structure on the pellet periphery (the rim) during both normal operations and under adverse transient conditions such as RIA. The remainder of the papers covered specific issues and are summarised below as miscellaneous items.

Thermally Activated Processes

The issue of intra-granular bubbles was the subject of two papers. L Noirot (CEA) considered the nucleation, growth, migration and re-solution of the small bubbles under irradiation conditions using a detailed finite-element scheme. Evans (UK) considered the problem of the behaviour of these bubbles in out-of-pile anneals in the absence of irradiation induced re-solution effects. He concluded that thermal re-solution of gas atoms from bubbles was not a physically viable mechanism and that release could only occur through bubble migration up the vacancy gradient emanating from the vacancy-rich grain boundaries. It is clear that much remains to be learned about this category of porosity since large swellings can result under transient conditions.

Two further papers considered the mechanistic interpretation of the *Vitanza Fission Gas Release Threshold* where new data indicate release at high burnups occurring at lower temperatures than expected. Van Uffelen (SCK/CEN) proposed the novel idea of athermal open porosity while Szuta (Poland) related the threshold to UO_2 grain growth and a *recrystallisation temperature*.

Schubert et al. (ITU) discussed Cs diffusion rates comparison of Cs and Xe profiles measured by EPMA. Using a simple method applied to an extensive database they demonstrated that Cs diffused at about 1/3 of the rate of Xe.

White (BNFL) reported detailed SEM measurements on ramped AGR fuel and demonstrated that the grain boundary bubble densities were consistent with continued bubble coalescence and that the final bubble morphologies depended critically on the relative rates of bubble growth and grain boundary and surface diffusion rates.

RIM/RIA

Baron (EDF) and a group of experts of ITU presented a comprehensive review of the rim structure, showing the radial variation and development of the thermal, transition and rim regions with burnup. Of particular interest were measurements of lattice parameter, micro-hardness, fracture toughness, porosity and Xe retention, performed at ITU during the last years. In this context, a long-term institutional program was running at ITU, aiming at the development of micro-characterisation techniques and at improving the knowledge of the high burn-up phenomena in the micro-scale range.

Desgranges (CEA) discussed the possibility that the *round* micro-grains surrounding large gas bubbles in the rim arose from a surface diffusion effect. The effect was not confined to spherical bubbles but was also observed in acicular porosity.

Lemoine (IPSN) considered the development of large swelling strains in small inter-granular bubbles in the rim region during RIA to result from the rapid adiabatic deposition of energy into the bubbles. The resulting stresses caused grain separation and subsequent gas venting. This idea was further developed by Muller (CEA) who predicted rapid oscillations in the resulting stresses.

J Noirot (CEA) examined the Kr release rates from fuel irradiated in the 900 MWe EDF stations to 5 cycles. By comparison with predictions for isotopic generation in various parts of the pellet he was able to demonstrate that in the low enriched fuel (3.1%) only about 1/3 of the release at 60 MWd/kgU originated from the rim. In contrast, in the higher enriched fuel (4.5%), which runs at higher temperatures, the contribution from the rim appeared to be negligible at these burnups, originating almost entirely from thermally activated processes in the pellet central regions. The analysis was extended to cover a number of rods in the CABRI series and the contribution to the release from the rim regions appeared minimal. This issue is clearly one of great importance for two reasons. From the normal operations point-of- view, the observation of the low contribution of the rim to high burnup gas release is of value while models for fuel failure under RIA generally predict gas venting from peripheral regions.

Miscellaneous Issues

Parashiv (Romania) considered the release of volatile species, particularly Te and I, to be limited by grain boundary diffusion effects through a polycrystalline medium. Validation of his highly detailed model was hampered by lack of suitable data.

Lewis (RMC-Canada) extended his defect fuel model to include the effects of high pressure steam and oxygen diffusion down the radial temperature gradients in the rod. Good agreement was found on a limited database using a Langmuir adsorption isotherm.

T Petit (CEA) considered the vacancy and interstitial formation energies in UO_2 using density function theory with muffin tin potentials. The work produced results in accord with expectations and is expected to be extended to MOX fuel.

Piron (CEA) presented a review of helium generation in stored irradiated UO_2 and MOX fuel. He demonstrated that helium generated by decay of actinides could present serious over-pressure problems for dry-storage, particularly for MOX fuel. Predictions of future behaviour were hampered by lack of knowledge of basic long-term behaviour, such as low temperature diffusion coefficients for helium.

The session included much lively discussion indicating the whole spectrum of agreement, partial agreement and total disagreement over many of the issues.

Summary of Session 3: Analytical Experiments

Chaired by: M. Tourasse (CEA) and E. Kolstad (Halden Project)

The first part of the session was focused on papers devoted to experimental techniques operating in hot cell laboratories. Session chaired by M. Tourasse

1. R.S. Dickson, R.F. O' Connor, Don D. Semeniuk (AECL) : Grain boundary inventories for Kr in Candu fuels
2. L. Caillot, G. Eminet, S. Ravel, R. Salot, E. Muller (CEA) : Adagio : Partition of grain boundary and matrix gas inventories : results obtained using the Adagio facility"

The AECL paper showed new results obtained in the Chalk River facility presented by P.H. Elder and D.S. Cox in the 1994 IAEA technical meeting. After the first publication several technical contacts between Canadian (AECL) and French Laboratories (Grenoble) led to complementary investigations on the Chalk River facility that are presented in the present paper.

The CEA paper is a presentation of a new facility operating in LAMA based on AECL technical approach adapted to PWR fuel. First results are discussed and several questions arose in the paper.

These new techniques clearly correspond to a need for a better understanding of fission gas location. The other classical techniques (sublimation, EPMA, XRF) do not answer the question concerning location of intragranular and intergranular gas. The work performed some years ago at AECL Laboratory showed the capability of the technique to answer the question.

Thanks to collaboration between Canadian and French scientists, we have now two facilities to perform such studies that allow fruitful comparison. The question of whether the extraction of gas is total at 1100°C or not arose 2 or 3 years ago at the beginning of the collaboration between both teams. The answer has been given by R.S. Dickson during the meeting: it is clear that the extraction is not complete (about half of the gas is not released!). The new facility ADAGIO in Grenoble induces the opening of a new question regarding the separation of both families of gases during the oxidation phase. Are ^{85}Kr small intragranular bubbles released during this process? The ADAGIO facility also confirms the opportunity of being very aware of the total extraction problem. Despite extractions performed at 1450 and 1750°C, the question remains open. During the meeting, a new question appeared, namely: are we sure that the pre-irradiation does not induce any modification of the gas distribution in the sample?

The future work has to be conducted in the following directions:

- continuation of the technical improvement of the technical process,
- investigation of the comparison between caesium release and gas release. Several discussions during the conference showed the interest of such comparisons (session 2),
- modelling of the phenomena of gas release during annealing needs to be clarified (we noticed some inconsistent approaches between the presentations made by S. Valin and J.H. Evans),
- the question of the fission gas release out of the rim region remained open even after the meeting. The ADAGIO technique could give a good answer to this question if sampling the rim region became possible or by using isotopic measurements of extracted gas.

The second part of the session was focused on papers devoted to experimental techniques operating in experimental reactors. Session chaired by E. Kolstad

1. S. Valin, A. Mocellin, G. Eminet (CEA), J.C. Joubert (INP-Grenoble): Modelling the behaviour of intergranular Fission Gas during out of pile annealing.

Standard UO_2 fuel irradiated in a commercial PWR (burn-up: 14 MWd/kgU) and hyper-stoichiometric large-grained fuel irradiated in an experimental reactor (burn-up: 9 MWd/kgU) were submitted to out-of pile annealing treatments under sweeping helium. The ^{85}Kr release was continuously monitored during the annealing experiments, with temperature ranging from 1400 to 1760°C. The burst release was not systematically observed during the heating ramp but sometimes during the isothermal period following the ramp.

A model based on microstructural observations was developed to explain the experimental results. The release was shown to depend on gas diffusion towards grain boundaries and on temporary gas trapping in grain boundary bubbles. The gas saturation on grain boundaries leading to release, depends on their crystallographic orientation.

Model calculations fitted well the experimental variations of ^{85}Kr release during anneals. The release mechanisms are the same for UO_2 and UO_{2+x} fuel.

2. J.A. Turnbull, E. Kolstad (Halden Project): Investigations on radioactive and stable fission gas release behaviour at the Halden Reactor.

Two types of experiments have been used in the Halden reactor to investigate the release of fission gases from LWR fuel. The first employs internal pressure sensors from which the kinetics and quantity of stable gases can be measured during irradiation. The second is the use of sweep gases to carry released fission gases from the fuel rod to a detector situated outside the reactor. With this equipment, it is possible to measure, using gamma spectroscopy, both radioactive and stable fission product release. In conjunction with fuel centreline thermocouples to measure fuel temperatures, these techniques have been successful in improving our understanding of the release process and the factors affecting it. The data generated have been used in many Member countries to develop models and validate fuel performance codes used in reactor safety assessments.

The phenomenon of stable fission gas release is now qualitatively well understood, and this has been achieved with the additional help of studies on the release of the short-lived radioactive species. The dominant processes are the single gas atom diffusion through the grains to grain boundaries and the interlinkage of porosity allowing escape into the rod free volume. In addition several other processes occur in parallel including intragranular bubble formation and irradiation induced resolution. There is also the possibility of release by other processes including grain boundary sweeping and bubble migration. Although it is possible to predict qualitatively the effect of parameters and design variables such as grain size, power and ramp conditions on release, there is less confidence in quantitative predictions. For this reason there is still a need for well-qualified data in order to refine existing calculational methods. In particular the threshold for release in the burn-up range above ~30 MWd/kgUO_2.

3. K. Malen: PIE/modelling of fission gas release from a Ringhals high BU rod bumped in Halden.

An ABB Atom fuel rod irradiated in Ringhals 1 (BWR) to a peak burn-up of 68 MWd/kgU has been refabricated at Kjeller and irradiated (bumped) in the Halden reactor. The fission gas release was 20%. The gas composition corresponds to the calculated composition of the generated fission gas at a burn-up of about 80-85 MWd/kgU. Ceramography and SEM investigations show that the release is from the central region, out to about 50-60% of the fuel radius. From thermocouple readings, Halden inferred a peak centreline temperature at axial maximum power position (solid pellet) close to 1350^{o}C. The fuel centre shows normal changes for this temperature and there is a rim zone about 150 micron wide. Code calculations were performed for estimation of temperature and FGR. Using burn-up corrected fuel thermal conductivity, it was possible to reach temperature and FGR values close to the measured ones - without the correction the calculated values were far too low.

Summary of Session 4: Industrial Modelling and Software Packages

Chaired by: K. Lassmann (ITU) and D. Baron (EdF)

Eight papers were presented in this session with emphasis on application, i.e. the usage of detailed (mechanistic) or simple (empirical) models within the analysis of fuel rod performance.

The paper of M. Tayal et al. showed the huge influence of local temperatures on fission gas release. A two-dimensional thermal analysis was performed that takes circumferential variations of gap size etc. into account. Comparing these temperature fields with axi-symmetric fields shows pronounced differences, which lead also to different fission gas release predictions. The authors come to the conclusion that this theoretically better thermal analysis significantly improves the predictions of fission gas release. In the discussion the remarkable agreement between calculated and measured fission gas release was discussed.

Two papers were related to purely empirical fission gas release models: the paper of N. Djourelov and the one of H.H. Lopez. There was agreement that such models are valid only for one specific design and a rather limited range of applications.

The paper L.C. Bernard et al. showed how relatively simple models with a clear physical meaning can be developed. Extensive verification was performed and it was outlined that industry needs models with good predictability that can easily applied, are reliable and fast running.

A very detailed (mechanistic) fission gas release model was presented (H. Wallin et al.) This model is incorporated in the fuel code SPHERE-3, which treats sphere-pac fuel. The model is based on several classical concepts and has been discussed in detail with experts (for instance T. Turnbull and R. White). Thus is must be considered as a model representing the state-of-the-art. However, verification has just started and the many parameters will be very difficult to be adjusted to the many different situations.

The status of the Russian code START-3 has been given in the paper of Y.K. Bibilashvili et al. The models of START-3 are a combination of mechanistic and empirical approaches. Physical details of the formation of the High Burnup Structure are discussed. The authors conclude that the formation of this structure near the rim has an impact on the temperature level in the fuel and is responsible for the observed enhanced fission gas release at high burnup. The authors claim that the release from the outer zones, in which more Pu is fissioned is consistent with the measured Xe-to-Kr ratio. Extensive numerical tests were performed to study the numerical behaviour of the START-3 code, which was found to be very satisfying.

K. Lassmann analysed numerical algorithms to solve the diffusion equation under varying conditions. Two algorithms were found to be superior, which may be obtained on request. The underlying assumption of a perfect-sink-boundary condition was controversially discussed.

C. Struzik reported a comparative study of fission gas behaviour in UO_2 and MOX fuel using the METEOR code. The obvious difference between UO_2 and MOX due to the different radial power density distribution is able to explain more or less the differences up to 50 MWd/kgM under normal conditions. However, the different behaviour in transients at a burnup above 30 MWd/kgM needs further clarification.

P. Chantoin et al. gave some first information on the Jules Horowitz Reactor which is a new research reactor dedicated to materials and nuclear fuel testing. The envisaged new on-line measurement techniques (e.g. on-line fission gas analysis, gap measurements, gamma spectroscopy, pressure (acoustic) measurements) were considered as extremely valuable for obtaining more detailed information.

Personal remarks K. Lassmann

1. Mechanistic models are indispensable and should remain a research tool also in the future. However, the consistent fitting of the many parameters involved (which may not even be parameters but further submodels) needs to be more transparent. Researchers must recognise that fitting of a large number of parameters (10-20) within a system of highly nonlinear equations, where most parameters cannot be observed directly, is impossible if only a very limited number of experimental results with large scatter is available. An independent assessment of mechanistic models seems to be necessary prior to the usage within fuel rod performance codes. Models from the eighties cannot be used without a complete refitting since at that time fuel temperatures were largely underestimated.
2. Most of the papers in this conference concentrated on intra- and intergranular fission gas release. Whether the transport along many grains of very different conditions (temperature, pressure etc.) can more or less be neglected (normally fast diffusion along grain boundaries is applied) is not sure.
3. Model constants fitted in a specific “environment” of models cannot automatically be used in different models. In general these constants are not principle constants but fitting parameters.

PROGRAMME

Tuesday, 26 September 2000

09.30 : Welcome **by Christian Bonnet**, Chairman of the meeting
09.45 : Opening Keynotes "Industry Challenges and Expectations with Respect to Fission Gas Release", *M. Lippens, D. Boulanger, L. Mertens (BELGONUCLEAIRE, Belgium)*
10.15 : Announcement on a IAEA co-ordinated activity on the subject, *P. Menut (IAEA)*

10.20 to 15.00 : **SESSION 1 : FEEDBACK FROM EXPERIENCE**
Session chaired by: M. Trotabas (COGEMA)
M. Billaux (SIEMENS CORP./USA)

- 10.20 : "Fission Gas Release and Related Behaviour of BWR Fuel under Steady and Transient Conditions", *H. Sakurai (NFD), M. Sasaki (JNF)), O. Kubota , T. Isogai (NUPEC)*
- 10.50 : "Fission Gas Release from High Burnup VVER-440 Fuel under Steady State and Transient Operation Conditions", *A. Smirnov, B. Kanashov, S. Kuzmin, F. Kryukov, I. Kungurtzev, G. Lyadov, (RIAR-Dimitrovgrad), A. Panyushkin, Eu. Bek (JSC "MSZ")*
- 11.20 : "Irradiation Test of DUPIC Fuel" *K.C. Song, M.S. Yang, H.S. Park (KAERI)*
- 11.50 : "Fission Gas Release and Fuel Swelling at Burn-ups Higher than 50MWd/kgU", *S. Brémier, R. Manzel (Siemens AG), C.T. Walker (CCE)*
- 12.20 : "Studies on Fission Gas Release Behaviour of BWR, and Experimental MOX Fuel Elements", , *U.K. Viswanathan, S. Anantharaman, D.N. Sah, S. Chatterjee, K.C. Sahoo, D.S.C. Purushotham (Bhabha Atomic Research Centre)*

12.50 to 14.30 : Lunch break

- 14.30 : "A Review of Fission Gas Release Data Within the NEA/IAEA IFPE Database", *J.A. Turnbull (Consultant), P. Menut (IAEA), E. Sartori (OECD/NEA)*

15.00 to 16.30 **: SESSION-2 : BASIC MECHANISMS (PART ONE)**

Session chaired by : R.J. White (BNFL)
J. Rouault (CEA)

- 15.00 : "A Mechanistic Fission Gas Behaviour Model for UO_2 and MOX Fuels", *L. Noirot, Ph. Garcia, C. Struzik, (CEA)*
- 15.30 : "Modelling the Influence of the Athermal Open Porosity on Fission Gas Release in LWR Fuel", *P. Van Uffelen (CEN/SCK-Mol)*

16.30 : Adjourn

Wednesday, 27 September 2000

09.00 to 18.00 : **SESSION-2 (CONTINUED) : BASIC MECHANISMS**

Session chaired by : R.J. White (BNFL)
J. Rouault (CEA)

- 09.00 : "Modelling of Fission Gas Bubble Migration to Grain Boundaries During Post-irradiation Annealing in High Burn-up UO_2", *J.H. Evans (Consultant*)
- 09.30 : "The Role of Grain Boundary Fission Gases in High Burn-up Fuel under Reactivity Initiated Accidents Conditions", *F. Lemoine, J. Papin, J.M. Frizonnet, B. Cazalis, H. Rigat (CEA)*
- 10.00 : "The Development of Grain Face Porosity in Irradiated Oxide Fuel", *R.J. White (BNFL)*

10.30 to 11.00 : Coffee break

- 11.00 : "Influence of Grain Growth Kinetics on Fission Gas Release", *M. Szuta (Inst. Of Atomi Energy-Poland)*
- 11.30 *:* "A Dynamic Model for the Fission Gas Induced Swelling in High Burn-up Fuel under Fast Power Transient", *E. Muller, F. Lemoine, R. Saurel (CEA)*
- 12.00 : "Contribution of the Rim to the Overall Fission Gas Release : What Do Isotopic Analyses Reveal ?", *J. Noirot , L. Desgranges, P. Marimbeau (CEA)*
- 12.30 : "Influence of Surface Rearrangement on Fission Gases diffusion : How Can Round Grains Contribute to the Fission Gas Depletion in the Rim", *L. Desgranges , N. Lozano, J. Noirot, B. Pasquet, G. Eminet (CEA)*

13.00 to 14.30 : Lunch break

- 14.30 : "Rim Formation and Fission Gas Behaviour: Some Structure Remarks", *J. Spino, D. Papaioannou, I. Ray (ITU), D. Baron (EDF)*
- 15.00 : "Study of the Atomic Rare Gas Behaviour by "Ab-Initio" Calculations", *T. Petit (CEA)*
- 15.30 : "Diffusion of Volatile Fission Products in Oxide Fuels Treated as Polycrystalline Materials", *M.C. Paraschiv, A. Paraschiv, V.V. Grecu" (Inst. For Nuclear Research, Pitesti-Romania)*

16.00 to 16.30 : Coffee break

- 16.30 : "Modelling of Fuel Oxidation Behaviour in Operating Defective Fuel Rods", *B.J. Lewis, B. Szpunar (Royal Military College-Ontario)*
- 17.00 : "Helium Behaviour in Spent UO_2 and MOX Fuels", *J.P. Piron, M. Pelletier, J. Pavageau (CEA)*
- 17.30 : "On the Diffusion Coefficient of Caesium in UO_2 Fuel*", K. Lassmann, A. Schubert J. Van de Laar, C.T. Walker*

18.00 : Adjourn

Thursday, 28 September 2000

09.00 to 12.30 **:** **SESSION-3 : ANALYTICAL EXPERIMENTS**

Session chaired by : M. Tourasse (CEA)
E. Kolstad (Halden Reactor Project)

- 09.00 : "Grain Boundary Inventories of Krypton in CANDU Fuels", *R.S. Dickson, R.F. O'Connor, D, D. Semeniuk (AECL)*
- 09.30 : Partition of Grain Boundary and Matrix Gas Inventories : Results Obtained Using the ADAGIO Facility", *S. Ravel, E. Muller, G. Eminet, L. Caillot (CEA)*

10.00 to 10.30 : Coffee break

- 10.30 : "Modelling the Behaviour of Intergranular Fission Gas During Out-of-pile Annealing", *S. Valin, A. Mocellin, G. Eminet, S. Ravel (CEA), J.C. Joubert (INP-Grenoble)*
- 11.00 : "Investigations on Radioactive and Stable Fission Gas Release Behaviour at the Halden Reactor", *J.A. Turnbull (Consultant), E. Kolstad (Halden Reactor Project)*
- 11.30 : "PIE modelling of Fission Gas Release from a Ringhals High Burn-up rod Bumped in Halden", *K. Malén (Studsvik Nuclear AB)*

12.30 to 1400 : Lunch break

14.00 to 18.00 **:** **SESSION-4 : INDUSTRIAL MODELLING AND SOFTWARE PACKAGES**

Session chaired by : K. Lassmann (ITU-Karlsruhe)
D. Baron (EDF)

- 14.00 : "Fission Gas Model of the Fuel Code SPHERE-3", *H. Wallin, L.A. Nordström, Ch. Hellwig (PSI)*
- 14.30 : "Development of the Fission Gas Behaviour Model in the START-3 Code and its Experimental Support", *Yu.K. Bibilashvili, A.V. Medvedev, G.A. Khvostov, S.M. Bogatyr, L. V. Korystine (SSC VNIINM.-Moscow)*
- 15.00 : "Evaluation of Thermal, Mechanical and Fission Gas Release Behaviour for BWR Fuel Rods with TETO ", *H. Hernandez-Lopez (Inst. Nat. de Invest. Nucleares)*
- 15.30 : "Mechanistic Modelling of Gaseous Fission Product Behaviour in UO_2 Fuel by RTOP Code", *V.D. Kanukova, O.V. Khoruzhii, S. Yu. Kourchatov, V.V. Likhanskii, L.V. Matveev (SRC TRINITI)*

16.00 to 16.30 : Coffee break

- 16.30 : "Framatome Analysis of Fission Gas Release and Related Topics", *L.C. Bernard, J. L. Jacoud, P. Vesco (FRAMATOME)*
- 17.00 : "Fission-Gas Release at Extended Burnups : Effect of Two-dimensional Heat Transfer", *M. Tayal, S.D. Yu, J.H.K. Lau (AECL)*
- 17.30 : "Macroscopic Fission Gas Release Model Applied to Russian Fuel", *N. Djourelov (Inst. Nucl. Research and Energy-Bulgaria)*

18.00 : ***Adjourn***

Friday, 29 September 2000

09.00 to 10.30 : **SESSION-4 : INDUSTRIAL MODELLING AND SOFTWARE PACKAGES (continued)**

Session chaired by : **K. Lassmann (ITU-Karlsruhe)**
D. Baron (EDF)

- 09.00 : "High Burnup Diffusional Model to Describe the Loss of He", *M. Landskron F. Sontheimer (Siemens AG), M.R. Billaux (Siemens power Corp.) (not presented)*
- 09.00 : "Numerical Algorithms for Intragranular Diffusional Fission Gas Release Incorporated in the Transuranus Code", *K. Lassmann (CEC - ITU)*
- 09.30 : "A Comparative Study of Fission Gas Behaviour in UO_2 and MOX Fuels Using the METEOR Fuel Performance Code", *C. Struzik , P. Garcia, L. Noirot, (CEA)*
- 10.00 : "The Jules Horowitz Reactor (JHR) : Experimental Possibilities", *P. Chantoin, L. Caillot (CEA), F. Augereau (LAIN UM2)*

10.10 to 10.30 : Coffee break

10.30 to 12.30 : **SESSION-5 : PANEL DISCUSSION : CONCLUSION AND PERSPECTIVES**

Session chaired by : **J.A. Turnbull (Consultant)**

Panel discussion board: **All the Chairs of the Technical Sessions**

12.30 to 14:00 : Lunch Break

14.00 to 16.00 : Visit to the "LECA" : CADARACHE PIE Facility

Seminar on Fission Gas Behaviour in Water Reactor Fuels

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