TRANSMUTATION: A DECADE OF REVIVAL ISSUES, RELEVANT EXPERIMENTS AND PERSPECTIVES

OVERVIEW PAPER

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Abstract

For more than a decade, transmutation studies have been again a topic of wide interest and have triggered numerous international activities, like bilateral/multilateral collaborations, information exchanges, state-of-the-art reports, conferences, but also some co-ordinated programmes and experiments.

It is legitimate to ask at this point, whether transmutation studies are still “fashionable” and why; what is known, what has been done and what should be done.

Since the motivations of national programmes are often different, due to a different context, we will take for granted that transmutation is generally seen as an option for the back-end of the fuel cycle in order to reduce the burden of potential geological storages of radioactive wastes (whatever their nature).

Finally, we also acknowledge the fact that some highly respected scientists have at several occasions during this decade expressed their doubts about the value of the transmutation option. A typical example is the position expressed by Pigford and Rasmussen, reporting the results of a study for the US National Research Council (IAEA-TECDOC-990, 2/12/1997).

1. Introduction

To give a state-of-the-art of the transmutation studies, one could make use of international publications or proceedings of the specialised conferences that have been mushrooming in this field. Of course a significant example is the OECD/NEA state-of-the-art report published in 1999: “Actinide and Fission Product Partitioning and Transmutation. Status and Assessment Report”.

We will limit our analysis to a few points that we consider of special relevance. Successively, we will review some ongoing research and experimental validation studies, in order to provide a list of relevant expected results, which should have impact to shape (or re-shape) future programmes.

In this perspective, we will also indicate which are in our opinion the “missing” experiments or studies, the absence of which could jeopardise the process of decision making.

The nuclear energy “environment” is a changing one, and it is of interest to review some activities/studies/concept proposals, which are not strictly speaking in the “transmutation” domain, but which could have an impact on the conclusions which could be drawn on the potential role of transmutation.

Finally we will attempt to summarise a list of open questions and an analysis of possible (re)orientation of priorities. Of course, this paper does not deal with chemistry issues, but rather with reactor and fuel cycle technology, since the physics of transmutation is today well understood (see for example [1]).

2. Where are we?

Transmutation is of course an R&D endeavour. The potential “customers” of such R&D can be found more in the society at large and its political representative bodies than in industry. By the way, it is not evident that even a fundamental feature of transmutation (i.e. the need to reprocess the fuel) is clearly understood in that context.

In so far as customers, one should not forget that utilities look probably with some apprehension and scepticism to studies which could offer options for the back-end of the fuel cycle but which could potentially have a non negligible impact on the cost of the electricity generation, without a clear definition of criteria to evaluate costs versus benefits, and in a frame of a highly competitive environment. In fact, utilities are ready to contribute to the R&D studies but to establish sound figures for induced or direct costs on the kWh!

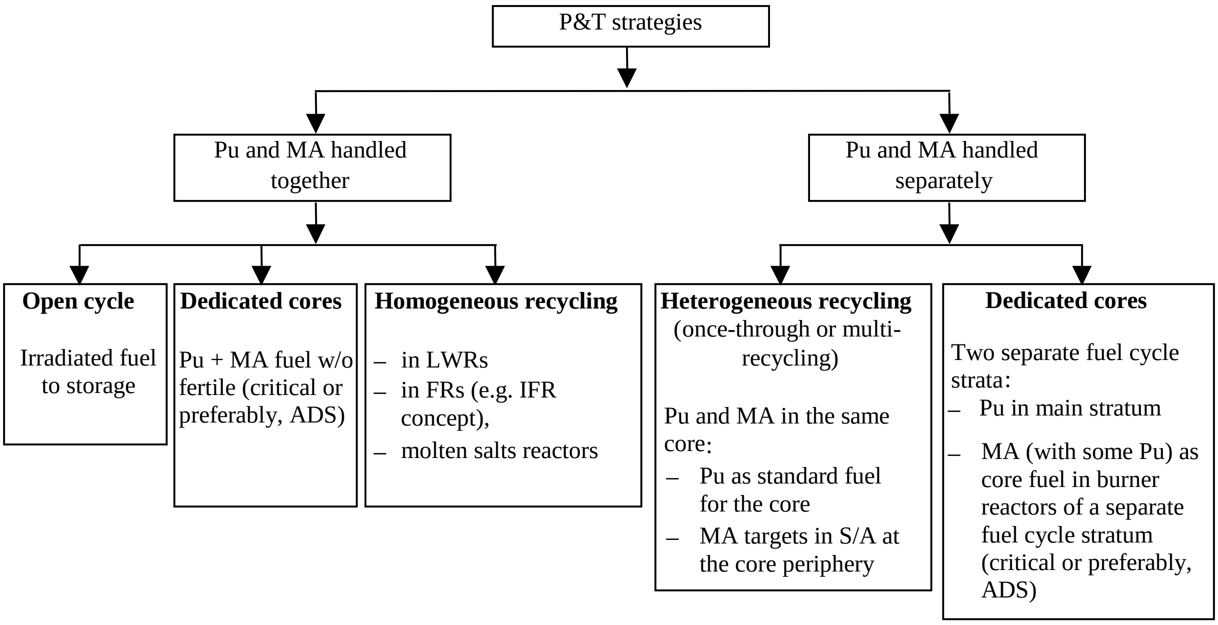
The nuclear reactor and fuel cycle industry has an obvious interest to be well informed of the transmutation R&D issues and results, but does not finance these activities other than marginally. This oversimplified analysis is only meant to make clear that there is an inherent difficulty to evaluate the real status of the research in the transmutation field with respect to the potential utilisation of the technology in a reasonable time horizon within stated performance and cost criteria.

In view of this difficulty, we have preferred to single out some specific scientific results, which could characterise our present understanding of transmutation and its use as an option for the fuel cycle.

2.1 *The IFR concept and the homogeneous recycling*

The IFR concept [2] is still the most outstanding example of an “inherently” transmutation concept in the so-called “homogeneous” recycling mode (see Figure 1 and [3]). The IFR concept can be seen as an energy producing system capable to recycle Pu and minor actinides (MA), to reach equilibrium, both stabilising the Pu and MA mass flows, and sending to the wastes only a very small fraction of the radiotoxic isotopes. This fraction is of the order of 0.1% or less, according to the announced performances of the pyrochemical process involved, which has still to be demonstrated at large scale in the frame of the transmutation application.

Figure 1. **Pu and MA management in P/T strategies**[6]



The appealing aspects of the IFR concept in the frame of transmutation are:

- The concept is mainly designed to produce energy, making an optimised use of resources and using a robust reactor and fuel cycle layout.
- The fuel cycle does not imply the separation of Pu and MA.
- The concept can accommodate in principle several options in terms of reactor size, reactor coolant, waste-forms, etc.

In general, the homogeneous recycling has equivalent performances for whatever the type of fuel in the fast reactor. In fact, if the losses at reprocessing are assumed to be of the order of 0.1%, the homogeneous recycling allows to reach a reduction of the potential radiotoxicity with respect to the open cycle scenario of a factor of 200 and more, and this over all the time scale ($10^2 \rightarrow 10^6$ years) [4]. However, the consequences on the fuel cycle have to be taken into account (see Table 1) and their impact evaluated.

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[6] If LLFP management is required, they can in principle be handled in the different scenarios as targets to be irradiated at the periphery of the different core types.

Table 1. **Consequences on the fuel cycle of MA recycling in FR[a].**
Variation expressed as ratio with respect to the corresponding values for the reference case: PWR-MOX Pu content: 12%, taken as 1.

		PWR – MOX 12%	**FR: Multi-recycling of Pu, Am, Cm**	**FR: Multi-recycling of Pu, once-through irradiation of Am + Cm targets[d]**
Fabrication	Activity heat due to: α β γ Neutron source	1 1 1 1 1	0.1 0.5 0.2 1.5 30	0.1 1.7 0.4 8.7 104
Reprocessing[b]	Activity heat due to: α β γ Neutron source	1 1 1 1 1	0.1 0.2 0.5 0.2 0.4	0.10 0.11 0.09 0.06 245[c]

a) Oxide fuel, EFR type.
b) 5 years cooling time.
c) Effect due mainly to ^{252}Cf.
d) Heterogeneous recycling total fission rate: 90% (see text).

2.2 Heterogeneous recycling and its potential limitations

An option has been explored, mainly in Europe and in particular at CEA in France [5] and at JNC in Japan [6], to perform the transmutation of MA in the form of targets to be loaded in critical cores of a "standard" type. The mode of recycling has been called "heterogeneous" (see Figure 1), the potential advantage being to concentrate in a specific fuel cycle the handling of a reduced inventory of MA (separated from Plutonium). The major obstacles to that approach are:

- The very high irradiation doses needed to fission a significant amount of MA (which implies very high damage rates).
- The need to separate Am and Cm from Pu and to keep them (Am and Cm) together, in order to reach high values (~30) for the radio-toxicity reduction [3].
- The need to load the MA targets in a very large fraction (~30 ÷ 50%) of the reactor park, possibly made of fast reactors, due to their favourable characteristics for this mode of recycling (high fluxes, which can be easily tailored in energy to increase fission rates).
- Consequences on the power distributions and their evolution with time.

In any case, the consequences on the fuel cycle are relevant, if one wants to reach a factor of radiotoxicity reduction of ~30 ÷ 40 (see Table 1 and [7]).

2.3 Dedicated systems

Making again reference to the scheme of Figure 1, a possible approach to keep the MA fuel cycle and the transmutation technology separated from the electricity production, is the one which calls for the use of "dedicated" cores, where the fuel is heavily (>30%) loaded with MA, the rest being, e.g. plutonium (the ratio Pu/(Pu + MA) being <0.3). Work performed at JAERI in Japan [8] and in Europe [9], has shown that critical "dedicated" cores can have difficulties, related to the safety parameters degradation, due in particular to a very low delayed neutron fraction (<0.2% $\Delta k/k$) according to the fraction of Pu (and its isotopic vector) in the core, and to a reduced Doppler effect. This characteristic has indirectly helped to promote the accelerator driven sub-critical systems (ADS) and the so-called "double strata" fuel cycle concept [8].

It should be said, however, that a convincing, scientifically based comparison of a critical and a corresponding (i.e. same fuel, same power, same coolant) sub-critical core is still lacking, in particular in terms of safety performances for a well defined range of MA/Pu ratios and for a variety of Pu isotopic vectors. As far as transmutation performances, obviously both critical and sub-critical systems (loaded with the same fuel and for the same power) are equivalent. The "dedicated" cores approach implies that ~3-5% of a power park (in other term a "support" ratio of ≈20) is needed to stabilise MA and Pu mass flows (see for example §3.3.3).

The "dedicated" core approach has been a powerful incentive to support ADS research studies.

It is also worth to notice, that the concept of a "dedicated" core applies both to a strategy of reduction mainly of MA or of reduction of Pu and MA (see Figure 1). In fact, Pu and MA can be kept together and their inventory reduced drastically, using a fuel without "fertile" (i.e. uranium) support. In this case (where the Pu/(Pu + MA) ratio is of the order of 0.7 ÷ 0.8), ADS can still offer a valuable core concept.

2.4 Long lived fission products (LLFP)

The transmutation of LLFP has always been a controversial issue. A better understanding of the list of potential candidates for transmutation has been achieved [10]. However, there is no doubt that the task of transmutation, even if limited to ^{99}Tc and ^{129}I (^{135}Cs being out of question) is a formidable one, in terms of fuel cycle requirements. In fact, the relatively slow pace of transmutation even in the most optimised neutron environment (i.e. high flux and well-tailored thermalized spectrum), implies the presence in the fuel cycle at equilibrium of a very large amount of these fission products [11]. The use of ADS can slightly help, in terms of neutron availability, but the decrease of the support ratio (unless ^{129}I isotopic separation is envisaged) makes all the scenarios of LLFP transmutation very unlikely.

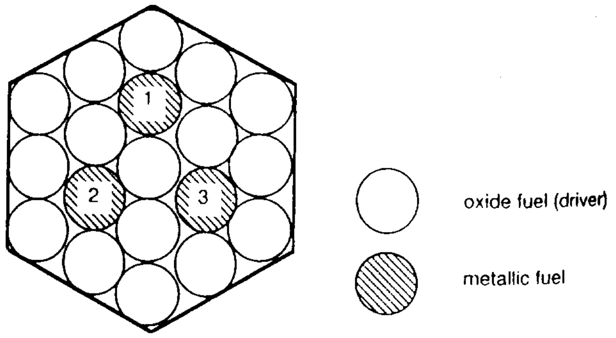
3. What is being done?

For the four areas indicated above, that we have chosen as relevant to assess the potential of the transmutation technology, it is important to review ongoing research and experimental validation studies, in order to evaluate the time horizon for their possible implementation.

3.1 The homogeneous recycling and the IFR concept

Unfortunately, after a period of intensive development, the IFR programme has been stopped. No significant activities have been left alive on the metal fuel development and the pyroprocess, with the exception of activities started at CRIEPI (Japan) and now extended to TUI-Karlsruhe. These new activities concern also the fuel reprocessing (see [12] at this conference). In particular, an experiment (METAPHIX) is planned, in order to irradiate metal fuel pins, loaded with MA and rare earths (RE) ([13] and Figure 2).

Figure 2. **Arrangement of fuel pins in a rig for the METAPHIX experiment (CRIEPI-TUI) (from [13])**



Nine metallic fuel pins are prepared for the METAPHIX irradiation study: three pins of UPuZr, three pins of UPuZr-MA2%-RE2%, three pins of UPuZr-MA5%, and UPuZr-MA5%-RE5%. They are planned to be inserted in the positions 1, 2 and 3, respectively, in the rig. Three rigs consisting of three sample metallic fuel pins and sixteen driver oxide pins will be prepared. Three rigs correspond to three different values of the burn-up: 1.5, 5 and >10%, respectively.

As far as homogeneous recycling in standard oxide fuels, some experimental knowledge has been obtained with the SUPERFACT experiment [14]. More will come from experimental programmes conceived at JNC and which should take place in JOYO beyond 2003. On the contrary, no experience exists on MA-loaded oxide fuels in standard light water reactors.

Finally, it has to be noted that the present revival of interest for pyroprocessing techniques, has been largely motivated by the relevance of these techniques to handle "hot" fuels, like those which are foreseen for transmutation. It has to be noticed that a modest but significant programme, PYROREP, has been launched as an EU contract for the 5th FWP.

3.2 Heterogeneous recycling

Apart from conceptual studies at JNC and CEA, experimental activities have been launched in Europe (e.g. the EFTTRA collaboration) and some useful indications have been gathered [15]. The EFTTRA-T4 and T4-bis experiments concern ^{241}Am, at a 12% volume fraction, inside a matrix of $MgAl_2O_4$, for a maximum fission rate of 28%. The swelling due to the decay of the ^{242}Cm produced by neutron capture, has been relevant, and triggered further research on the form of inert matrix/actinide fabrication (e.g. micro-dispersion versus macro-dispersion). It is worth to notice that experiments performed up to now, did not cover the presence of ^{243}Am and the presence of Cm.

Further experiments are planned in France, and in particular the ECRIX experiment, which should take place in PHENIX and the CAMIX and COCHIX experiments (J.C. Garnier, CEA, Private communication), also planned in PHENIX. The CAMIX experiment will provide information on "micro-dispersion" of a (Am, Zr, Y)O_{2-x} compound in MgO and COCHIX information on the same compound “macro-dispersed” in MgO or $(Zr_{0.6} Y_{0.4})O_{1.8}$. All 3 experiments are planned to reach a fission rate equivalent to 30 at%.

A significant global experiment is presently worked-out in the frame of the collaboration between MINATOM (Russia) and CEA (France) with the participation of FZK and TUI-Karlsruhe, as partners of CEA. In this experiment (AMBOINE), Am targets AmO_2+UO_2 and AmO_2+MgO should be fabricated at RIAR according to the VIPAC process. These targets should be irradiated in BOR-60 and reprocessed by pyroprocess after irradiation again at RIAR, providing in that way a full validation of the whole fabrication – irradiation – reprocessing cycle for CERCER targets (S. Pillon, CEA – Private communication).

3.3 Dedicated systems

3.3.1 Fuels for dedicated systems

For both critical and sub-critical dedicated cores, the major issue in the path towards feasibility demonstration, is the fuel development. Many candidates have been considered (see for example Table 2), but limited experimental work has been done, in order to characterise the basic properties of these potential fuels, their fabrication processes and their behaviour under irradiation.

Table 2. **Dedicated Pu + MA fuels (adapted from [16])**

Metal fuels	– Need to improve thermal properties ⇒ add non-fissile metal with high melting point (e.g. Zr) ⇒ Pu-MA-Zr alloy. – However: mutual solubility of Np and Zr?
Oxide fuels	– Mixed transmutation oxides as a logical extension of MOX. – However: smaller margin to melting (low thermal conductivity).
Nitride fuels	– Good thermal behaviour. – However: need to enrich in ^{15}N. – Lower stability against decomposition at high temperatures.
Composite fuels: the role of Zr	Ad-hoc “tailoring”: – MgO + (Zr, An)O_{2-X} (CERAMIC-CERAMIC). – Zr + (Zr, An)O_{2-X} (CERAMIC-METALLIC). – Zr + (An, Zr) alloy (METAL-METAL). However, fabrication can be difficult (also: size and distribution of the disperse actinide phase).
Coated particle fuels	Special form of composite fuels. However in the case of fast spectra, little is known on potential candidates (TiN?). ⇒ A generic problem: the high He production under irradiation.

A common feature for these fuels is to be fertile-free, or, at least, “U-free”, since Th is sometimes considered as an acceptable support, in particular for strategies that promote the replacement of the U-cycle with the Th-cycle.

Well-structured programmes for fuel development are missing in practically all the major transmutation programmes. A significant exception is the JAERI programme, focused on nitride fuels.

The CONFIRM project, sponsored by the EU 5th FWP is also devoted to nitride fuels ((Pu, Zr) N and (Am, Zr) N). The project aims to the fabrication, characterisation and irradiation of these fuels, and addresses also the issue of ^{15}N enrichment.

Finally, we recall the initiative of the European Technical Working Group (TWG) on ADS, chaired by Professor Rubbia, which has set up an ad-hoc task force an Fuel Fabrication and Processing, in order to produce a state of the art report and an agreed work plan in the frame of a road-mapping towards ADS deployment [16].

As far as reprocessing, the situation is, obviously, not fully satisfactory either. The use of dedicated fuels imposes their reprocessing in all considering schemes. Again, the programme proposed by JAERI, includes laboratory scale experiments of reprocessing, together with a flow diagram of a process to enrich in ^{15}N the fuel [17].

The present European efforts are reviewed in [12,16]. A programme for pyrochemistry development is also being set up in France at CEA. The rationale for it can be found in the report: "Assessment of Pyrochemical Processes for Separation/Transmutation Strategies: Proposed Areas of Research – CEA/PG – DRRV/Dir/00-92, March 2000".

3.3.2 ADS systems

A special case is the research activity in the ADS domain.

In the last two years, relevant initiatives have taken place. The ATW Roadmapping in the US [18], should give rise to a focused programme in very near future. The joint KEK-JAERI project, has given a place to ADS development in Japan, in the frame of a multipurpose facility [19].

In France, the GEDEON programme [20] has gathered a large community of physicists around the basic physics items of research for ADS (nuclear data, spallation physics, sub-critical core neutronics, materials, but also pyrochemistry, molten salts, thorium and system studies).

In Europe the Technical Working group mentioned above has been established. In that frame two concepts for ADS are being studied (see Figure 3) and a rationale is emerging for a "step-by-step" validation and demonstration of the ADS concept (see Figure 4) and its waste transmutation potential, in the frame of a specific road-mapping, which is being finalised at present. Few comments will be made in what follows, on some ongoing experimental steps, like the MEGAPIE project and the MUSE programme. Finally, the European Union is supporting a number of projects, in the frame of the 5th R&D Framework Programme.

The issues and programmes related to high power proton accelerators (HPPA), although essential, will not be dealt with here. We only remind the R&D work that has been initiated on the topic of "accelerator reliability" and which has been the subject of two NEA Nuclear Science Committee workshops (Mito, 1998, Aix-en-Provence, 1999).

Figure 3. **Sketch of ADS, liquid metal cooled (right) and gas cooled (left) (not to scale), representative of the European EADS proposals (ANSALDO and FRAMATOME). Potentially the same fuel assembly (e.g. SNR-300 S/A with MOX fuel).**

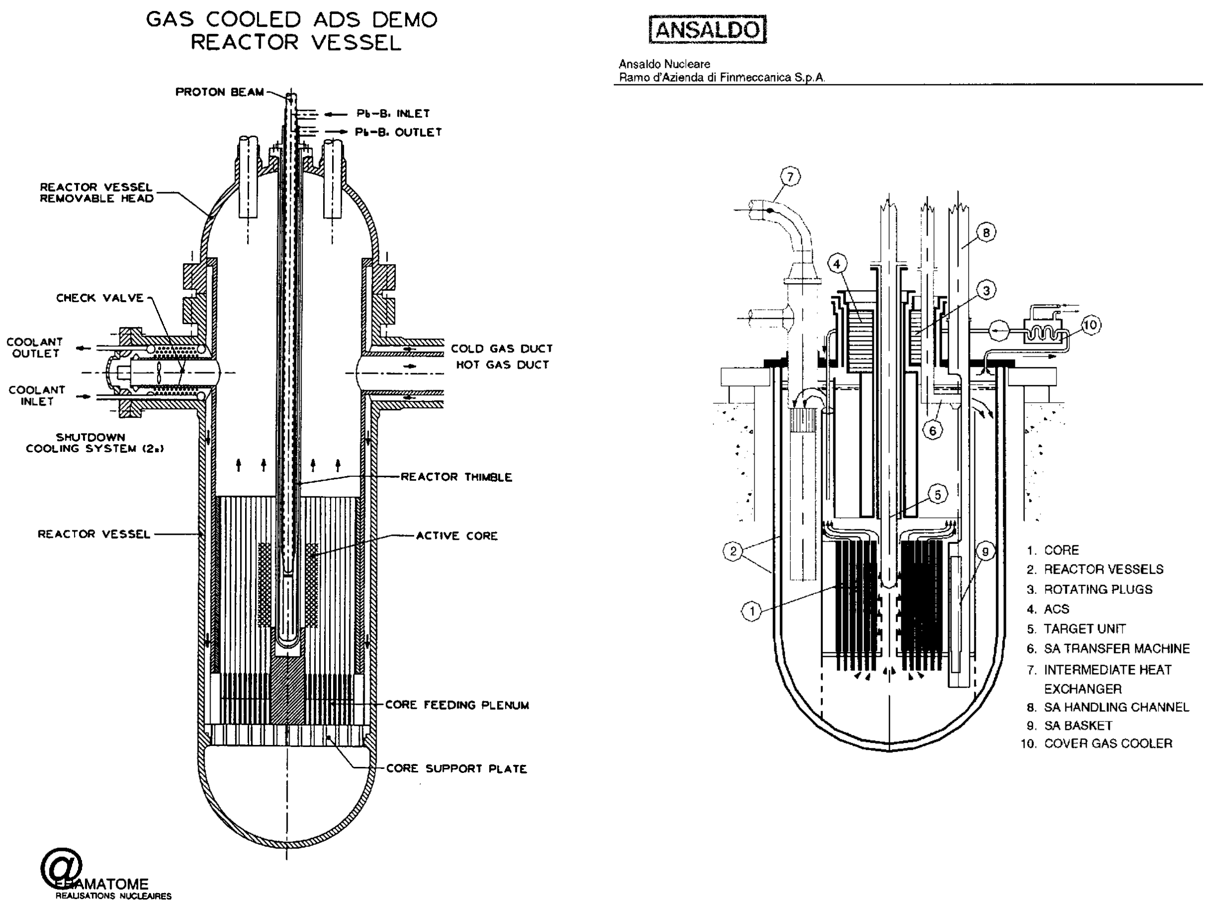
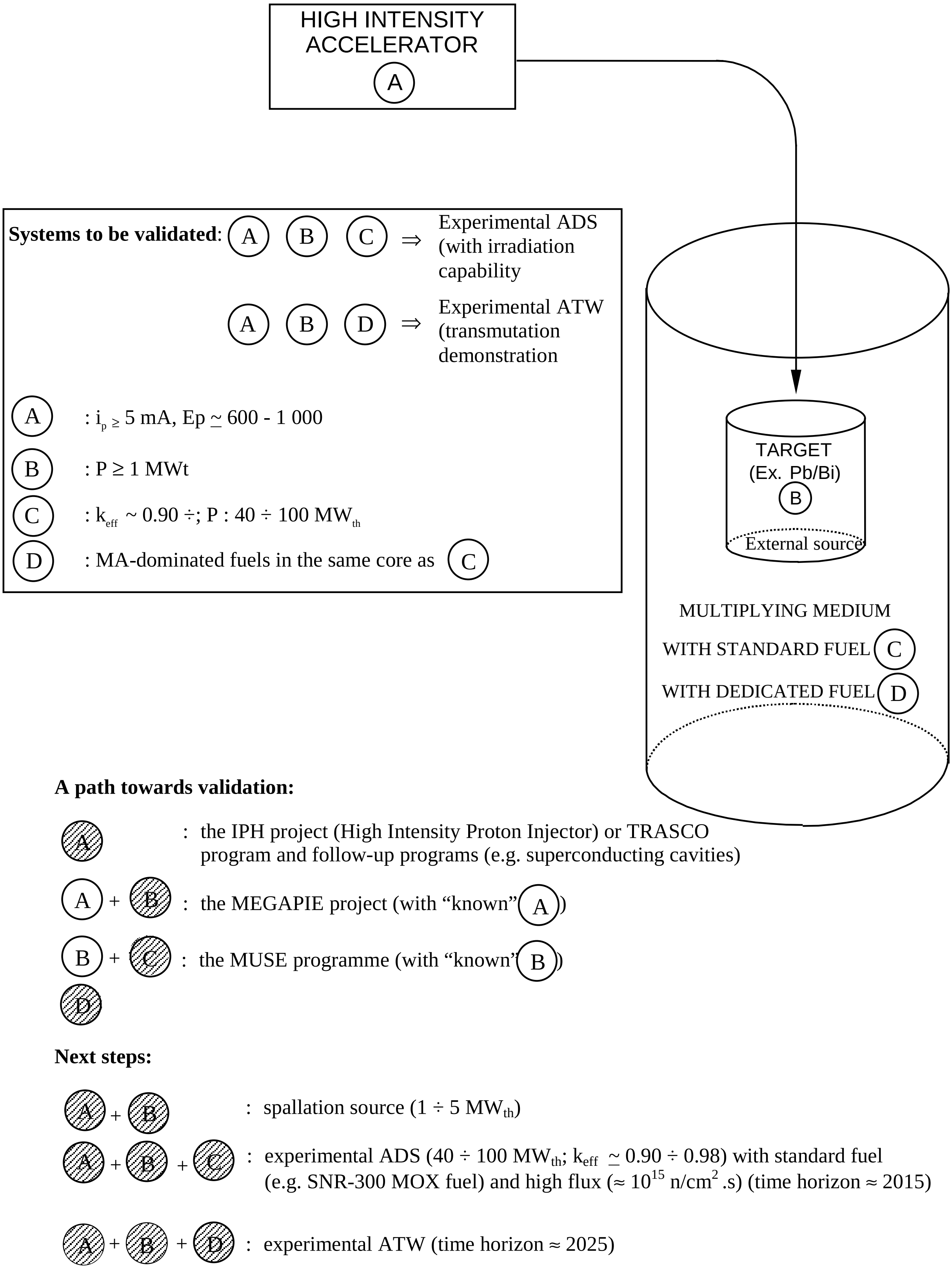


Figure 4. **A step-by-step approach to the validation and demonstration of the ADS concept**



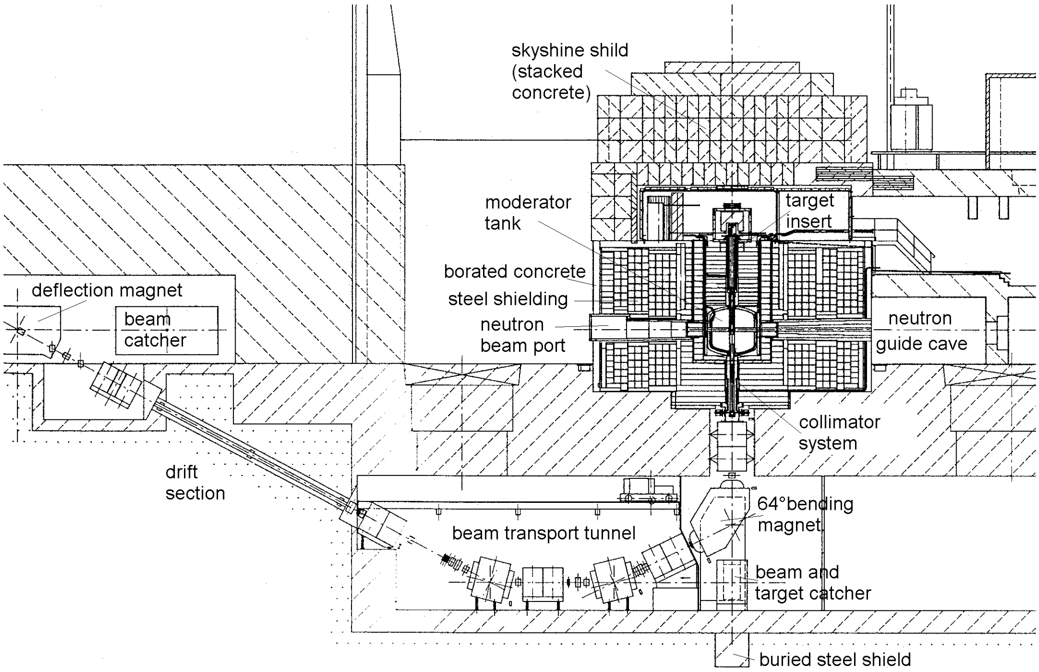
3.3.2.1 The MEGAPIE project [40]

MEGAPIE is an international (CEA, PSI, FZK, CNRS France, ENEA, SCK•CEN. JAERI will join soon) experiment to be carried out in the SINQ target location at the Paul Scherrer Institute in Switzerland and aims at demonstrating the safe operation of a liquid metal target at a beam power in the region of 1 MW. The minimum design service life will be 1 year (6 000 mAh).

The target material will be the PbBi eutectic mixture. Existing facilities and equipment at PSI will be used to the largest possible extent. In fact, the MEGAPIE target will be used in the existing target block of SINQ.

A vertical cut through this target block and parts of the proton beam line is shown in Figure 5.

Figure 5. **Vertical cut through the target block and part of the proton beam transport line of SINQ**



The target's outer dimensions must be such that it fits into the target position of the SINQ facility, the existing target exchange flask including its contamination protection devices and the existing target storage positions.

The target will be designed for 1 MW of beam power at a proton energy of 575 MeV, i.e. a total beam current of i_p = 1.74 mA.

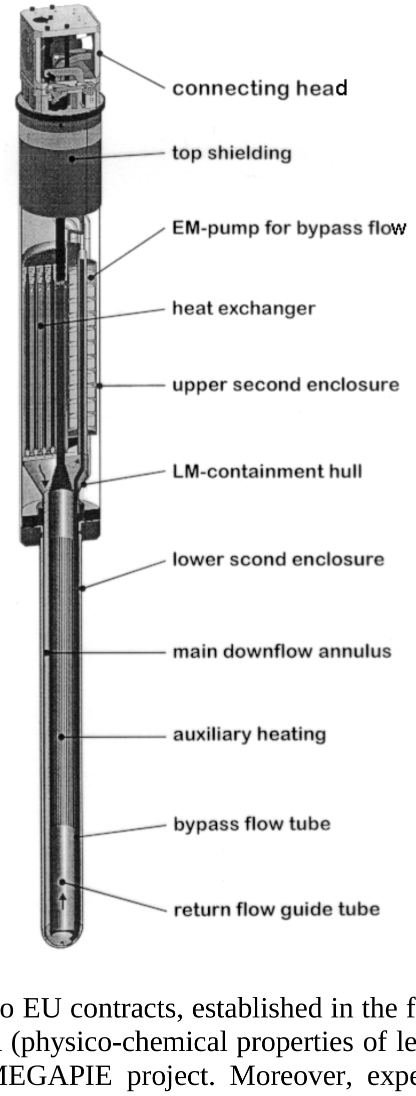
It is also important to realise that the stability of beam delivery cannot be guaranteed at all times. The MEGAPIE heat removal system must be able to cope with frequent short beam trips and occasional unstable operation i.e. up to days long shutdown periods.

A sketch of the MEGAPIE spallation target is given in Figure 6.

The major objectives of the MEGAPIE initiative are:

- Full feasibility demonstration of a spallation target system.
- Evaluation of radiation and damage effects of structures and beam window in a realistic spallation spectrum.
- Effectiveness of the window cooling under realistic conditions.
- Liquid metal/metal interactions under radiation and stress.
- Post irradiation examinations (PIE).
- Demonstration of decommissioning.

Figure 6. **Sketch of the 1 MW exploratory liquid lead-bismuth spallation target MEGAPIE**



It has to be reminded that two EU contracts, established in the frame of the 5^{th} FWP [21], SPIRE (material irradiation) and TECLA (physico-chemical properties of lead alloys: corrosion...), provide a relevant R&D back-up to the MEGAPIE project. Moreover, experimental laboratories have been launched in support of these activities (like the KALLA laboratory in FZK-Karlsruhe) or are re-oriented (like the ENEA laboratory in Brasimone: the CIRCE loop).

3.3.2.2 The MUSE experiments

The MUSE experiments, launched in 1995 [22], provide a simulation of the neutronics of a source-driven sub-critical system, using the physics characteristics of the separation of the effects due to the presence of an external neutron source from the effects of the neutron multiplication. In fact for a wide range of sub-criticality values (e.g. k_{eff}: 0.9 ÷ 0.99) the space dependence of the energy distribution of the source neutrons is quickly (in approximately one mean free path) replaced by the fission-dominated neutron energy distribution.

In practice, external known neutron sources have been introduced at the centre of a sub-critical configuration in the MASURCA reactor. The more recent of these experiments is made of a deuton accelerator and a target (deuterium or tritium) at the centre of a configuration, where actual target materials (like lead) are loaded, to provide the neutron diffusion representative of an actual spallation target (see Figure 7 and [23]). The neutrons issued from (d,d) and (d,t) reactions provide a reasonable simulation of the spallation neutrons, in terms of energy distribution (see Figure 8).

Static (e.g. flux distributions, spectrum indexes, importance of source neutrons) and kinetic parameters (e.g. time dependence of neutron population, effective delayed neutron fraction, with appropriate weighting, etc.) have been or will be measured (see [23]). Sub-criticality itself, is measured by static and dynamic techniques.

Finally, the proposed experiment MUSE-4 start-up procedure i.e. 1) critical configuration with accelerator hole but no beam, 2) sub-critical configuration with accelerator hole but no beam, 3) same, but with beam on, allows to establish a precise reactivity scale in step 1, which can be used both to calibrate eventual control rods and to measure in a standard way (e.g. with the modified source multiplication, MSM, method) the level of sub-criticality of steps 2 and 3.

The MUSE-4 experiments, described in a separate paper at this conference [23], are also partly supported by an EU contract for the 5th FWP.

3.3.2.3 Streamlining basic physics experiments

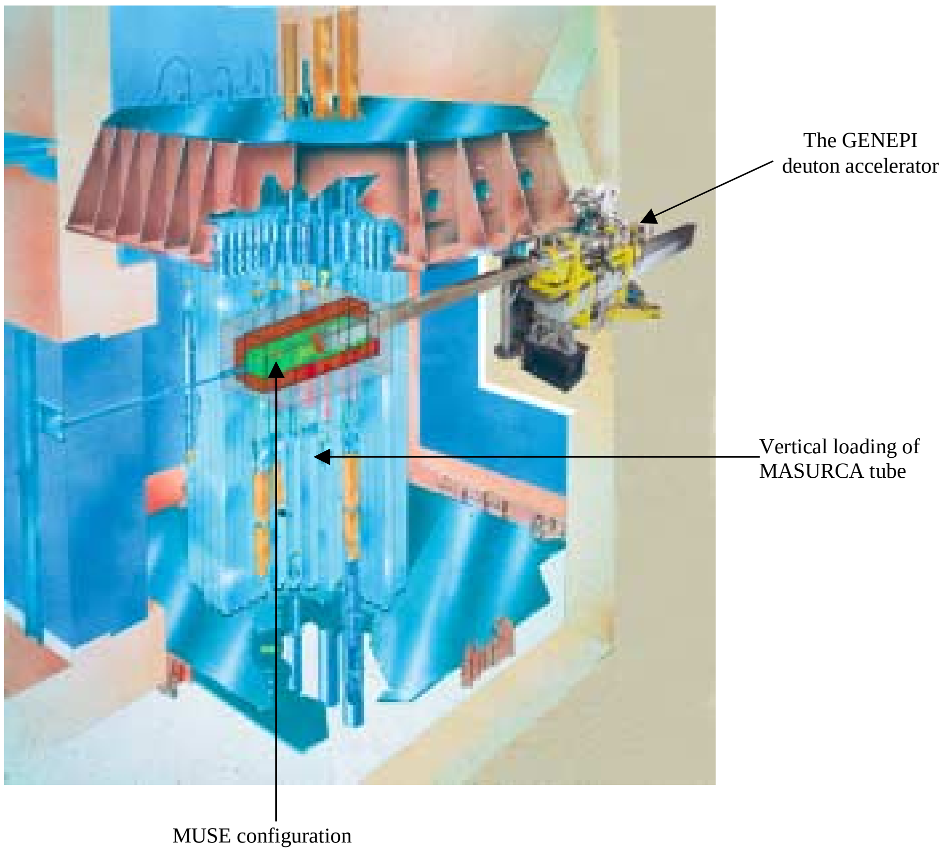
The ADS research development has also motivated a significant number of experimental activities in the field of spallation physics and nuclear data measurement and evaluation (mainly actinides and LLFP). Examples will be found in papers at this conference.

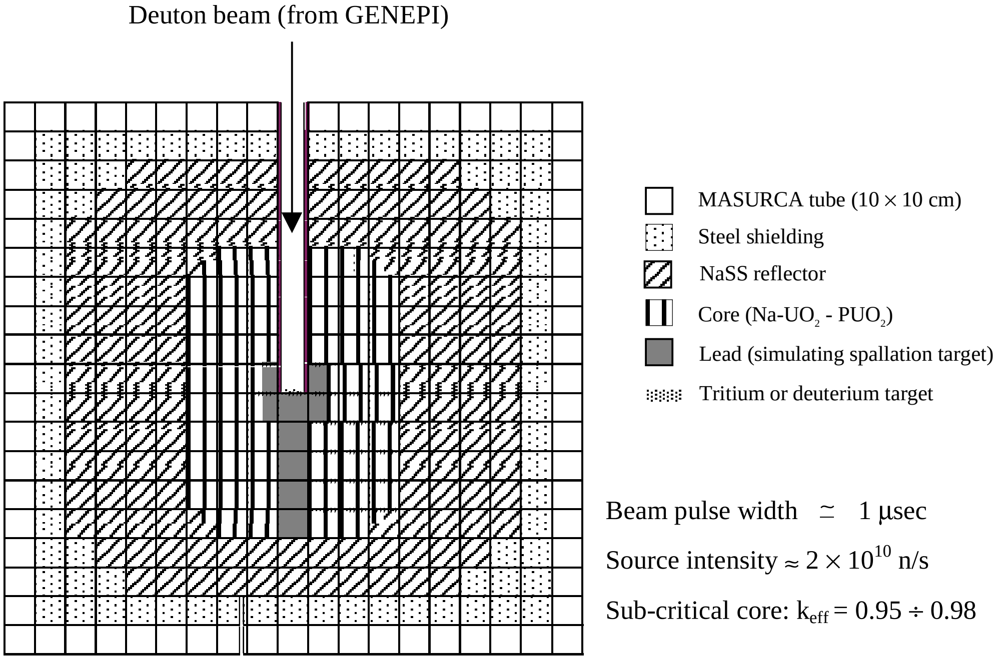
A major experiment takes place at GSI, defined in order to gather much needed information on spallation product yields and distributions in (A, Z) [24].

Also in this area, the EU supports projects in the frame of contracts for the 5th FWP [21].

If present uncertainties in nuclear data allow making reasonable pre-conceptual design assessments, future detailed studies will require more accurate data, with drastically reduced uncertainties. The relevant sensitivity studies have started (see [25]), but they have not yet tackled in satisfactory way the problem of the accuracy needs in the intermediate (i.e. 20 MeV ≤ E ≤ 200 MeV) energy range.

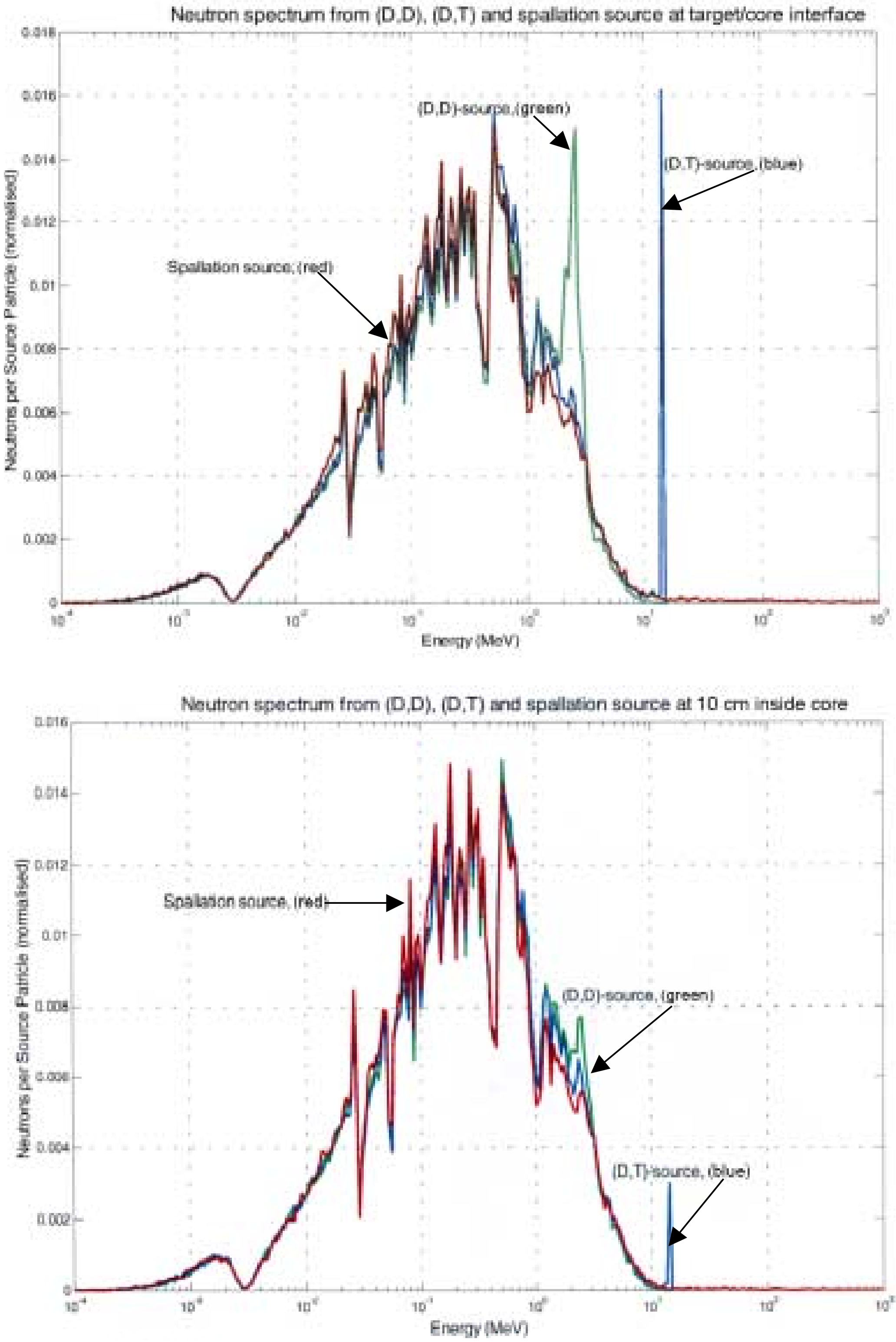
Figure 7. **The MASURCA installation for the MUSE programme**





A sub-critical MUSE-4 configuration

Figure 8. **Comparison of the neutron spectra obtained with (D,D) and (D,T) neutron sources (as in MUSE experiments), with the reference spallation source, in the same configuration**



3.3.3 Scenarios studies

Scenario studies have allowed during this decade to get a global picture of the transmutation potential, mass flows at equilibrium, and consequences on the power park structure. In the illustration of Figure 9 [26], the same type of ADS is used in order to transmute MA (double strata approach), or Pu + MA (double component type of power park [27]).

The fractions of ADS in the park at equilibrium are shown (respectively 3.4% and 16%), and also the MA and Pu yearly mass flows, including total losses towards a deep geological storage.

3.4 LLFP

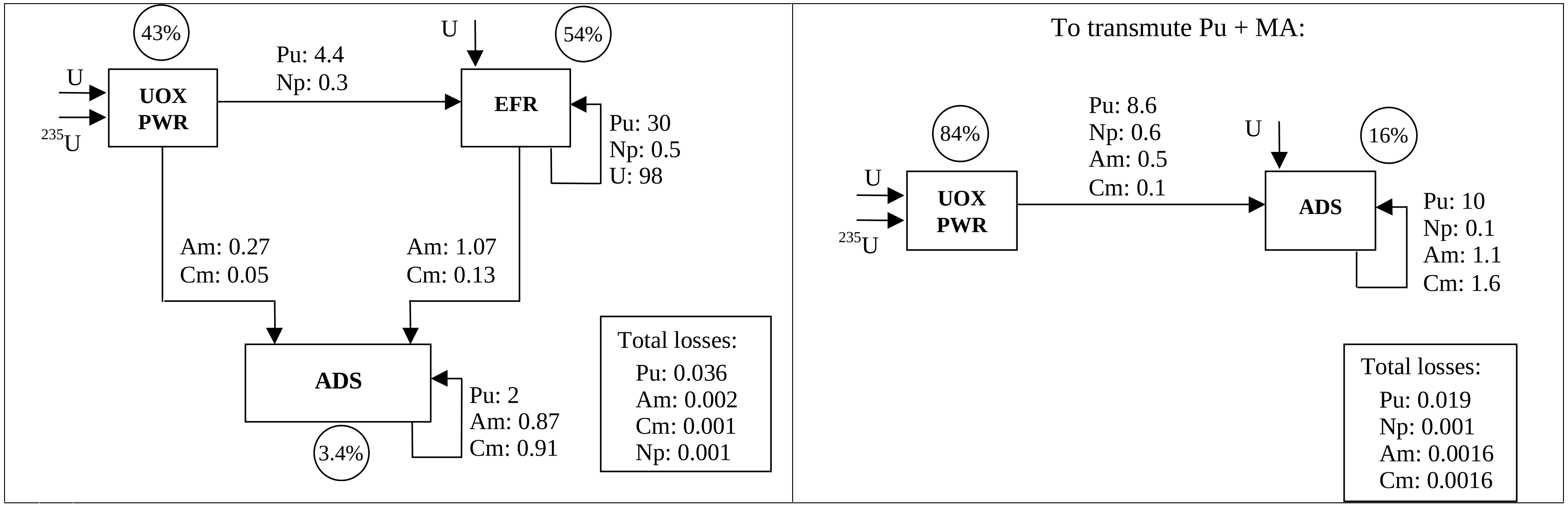
In this area, after the performance of irradiation experiments on ^{99}Tc and Iodine [28], not much is being done apart from conceptual studies, that underline the need to use high fluxes and thermalised spectra, like in the so-called "Leakage-with-Slowing Down" (LSD) approach [11].

Projects related to the transmutation of ^{90}Sr and ^{137}Cs have finally been abandoned everywhere.

Cs transmutation is more and more considered as non-realistic (even with isotopic separation). Other activation products have been mentioned as candidates for transmutation, but the inherent difficulty of high neutron-consuming processes has discouraged further experimental programmes.

Figure 9. **Scenarios at equilibrium for a 60 GWe Park – Mass flows/year (t) (only TRU)**

Double strata: to transmute MA (separated from Pu).
Double component: to transmute Pu + MA (non-separated).
Same type of sub-critical ADS (gas-cooled, particle-fuel).
Power: 1 500 MW_{th}.
Initial sub-criticality: $k_{eff} = 0.98$ ($i_p \approx 17$ mA, $E_p = 1$ GeV).



% : fraction in the power park.
UOX-PWR: 4.9% ^{235}U enrichment. BU: 60 GWd/t.
Cooling time: 5 years.
Ageing before irradiation: 2 years.
Losses to the wastes: U, Pu, MA= 0.1%.

4. Major missing points

The analysis of the ongoing activities and of their relevance allows attempting an indication of areas where further efforts are needed, in order to consolidate the present knowledge and to provide elements to judge feasibility.

- It seems that what is probably still needed with a high priority is a (re-)assessment of the criteria to judge the performance of transmutation systems. Transmutation as a waste management option, is indissolubly related to a constant or expanded use of nuclear energy and its impact should be evaluated on the full fuel cycle (cost and licensing of new installations, doses to the workers, secondary wastes, acceptability, ...).
- Experimentation about fuels is a priority. No concept can be considered seriously, if the appropriate fuels are not defined, which means characterised, fabricated, irradiated and reprocessed. Now, very limited facilities are available to deal with MA fuel fabrication and reprocessing (wet or dry routes) and their workload is already very demanding. Moreover, the problem of Cm has been up to now somewhat "forgotten" and, on the contrary, it can be crucial to define an optimum transmutation strategy or even to identify potential "show stoppers".

  To start with compounds and fuel basic properties assessment should be a priority. An international co-ordination and share of work should be envisaged.

  Also, irradiation tools with fast neutrons will be dramatically reduced in the coming years with the remarkable exception of the JOYO reactor (at least up to ~2015). Again, an international initiative could be envisaged to harmonise programmes and to allow the best use of existing resources.

  Reprocessing of irradiated fuels should be foreseen as an essential step of any programme on fuels, for homogeneous recycling of fuels, heterogeneous recycling of targets, or dedicated fuels. The priority, in the opinion of the author is with fuels for homogeneous recycling and with dedicated fuels, if the double strata, (or the "double component") approach is accepted.
- In the case of heterogeneous recycling, the feasibility of a fission rate >90% should be experimentally verified, and that demonstration should be made in the case of a target containing both Am and Cm. In fact, multi-recycling of targets should be avoided.
- It is more and more evident that transmutation studies could not necessarily require the separation of individual MA and MA from Pu. Quite the opposite option seems to be more attractive [3]. In that respect, partitioning/conditioning strategy [29] could represent an option to be investigated, and which could justify partitioning by itself.
- In the case of ADS, an Experimental ADS (EADS) realisation at the 2015 horizon, is a need, in order to prove the technology at a significant scale. The priority is the engineering concept (i.e. component coupling, control, reliability and licensing) validation. The double role of an EADS as a facility to validate the concept but also able to provide the appropriate fast neutron field for advanced fuel irradiation at high damage rates, is a strong point to be made.

  The present status and the necessary research in the accelerator field are the subject of another paper at this meeting. It is however necessary to remind here that this is an essential issue, since the expected performances of the "dedicated" high power accelerators are very demanding in terms of reliability and availability.
- LLFP transmutation research, if at all needed, should concentrate on a realistic approach for ^{129}I handling, if any. Reliable target materials and high transmutation rates should be the priority

goals in this field. Since once-through transmutation is hard to be envisaged, the recovery of iodine in the irradiated target and its reprocessing, should also be the object of research.

5. A changing environment

The research in transmutation experienced a revival in the mid-eighties, essentially in the context of waste management within programmes which gave a definite value to Plutonium and which implied the reprocessing of irradiated fuel.

The transmutation approach was successively identified in some countries with the approach to Pu elimination (both weapon and civil Pu). Reactor physics problems were indeed very similar. In that way, the interests of the two previously separated communities (i.e. Pu = resource versus Pu = liability), were somewhat federated, in particular in terms of fuels development and their reprocessing by pyroprocesses.

The third step in the evolution of the transmutation approach is underway at present, since the objectives of a "Generation IV" or, in general, the objectives of a future nuclear power development (beyond the horizon 2030-2050) are being globally re-discussed.

Transmutation and waste minimisation are then part of the potential criteria to define future energy systems (reactor plus fuel cycle).

In this changing environment, it can be useful to single out some concepts or research areas, which can have impact on the future of transmutation studies.

5.1 Evolutionary reactor concepts

A few well worked-out reactor concepts have emerged in the last few years, which, besides attractive safety and economics characteristics, have a potential to be "inherent" MA transmuters in the homogeneous recycling mode.

Besides the IFR concept, often quoted as a paradigm in the present paper and the Energy Amplifier proposed by C. Rubbia, we can remind the BREST lead-cooled fast reactor concept developed in Russia [30], the CAPRA reactor in France [31], the SCR (Super-critical Water Cooled) concept, developed in Japan [32], but also the APA concept [33], despite the fact that it concerns mostly an innovative assembly design for PWRs.

In particular, the nitride fuel foreseen for the BREST reactor, favours the MA transmutation by neutron spectrum hardening. Using a pyrochemical process, it is possible to envisage for this fuel, by multi-recycling, the transmutation of the actinide produced during irradiation. This mode, close to the one indicated for the IFR concept, has the same advantages and of course similar drawbacks, in particular due to the build-up of spontaneous fission neutron emitters (Cm isotopes, cf. isotopes, see §2.1 and Table 1).

Finally, interest in gas-cooled fast reactors has been renewed, in particular to keep open the fast reactor (FR) option, due to FR flexibility with respect to resources utilisation and their potential for waste minimisation. In view of the "political" opposition to Na as coolant in some countries, the gas cooling is being revisited.

The potential of any fast reactor, whatever the fuel type (oxide, nitride, metal) and whatever the coolant [34], indicates that the priority for GCFRs is to design a viable reactor (in terms of safety) with a realistic fuel form (e.g. particle, avoiding as far as possible graphite) for which no firm candidate has been proposed up to now.

5.2 Molten salts

Molten salt reactors, besides their specific interest as energy producing systems, have also a number of perceived advantages for transmutation, in particular:

- High burn-up potential (up to 600 MWd/t) limited only by absorption due to fission products, minimising the quantity of fuel to be reprocessed (a few litres of salt per day).
- Actinide losses minimised in the ultimate waste form (0.01-1% of the actinide inventory).

Molten salts characteristics result also in increased flexibility:

- Continuous input of purified salt fuel and output of irradiated salt: by adjusting the TRU concentrations, reactivity can be controlled without the use of poison or fertile material.
- Long-lived fission products (Zr, I, Tc, Cs, ...) can be added directly to the salt with no detrimental effect on its physicochemical properties.
- In the case of thorium cycle, fertile thorium can be added to the salt to fabricate ^{233}U rather than TRUs; the ^{233}U can be "quickly" extracted without ^{232}U.

Recent studies on molten salts concepts [35,36] point out the application to MA and Pu elimination, but also indicate the way towards improved fuel cycle and waste management scenarios (e.g. the TASSE system [37]).

5.3 The thorium cycle

A recent study of the European Union (in the frame of a contract for the completed 4th FWP, to be continued in the 5th FWP) has addressed the issue of "Thorium as a Waste Management Option" [38].

The objective of the work was a re-assessment of Thorium cycles in the context of limitation of nuclear waste production and prospects for waste burning. The aim was to obtain a review of the major steps of the fuel cycle, focusing to the waste aspect. A restriction was made regarding reactor types: PWR, FR and ADS.

The final report of that study shows that there are important advantages of thorium cycles with respect to the waste issue that we will quote in detail from [38]:

- Long-lived radio-toxicity of mining waste is expected to be relatively small, which leads to more manageable waste as compared to the uranium case.
- Fabrication of Th/Pu-MOX fuels is comparable with U/Pu-MOX fabrication methods as long as fresh Th, fresh U and recycled Pu are used. Recycling of U bred from Th, however, needs remote handling and reprocessing techniques specific to Thorium.

- The use of Thorium in PWRs always requires make-up fuel and therefore a self-sustaining mode is impossible in such a reactor.
- To reduce the radio-toxicity of PWR waste in an once-through mode, one has to avoid ^{238}U and therefore use thorium together with make-up fuel like ^{233}U or highly-enriched ^{235}U. Advantages in terms of waste radio-toxicity are seen during the first 10 000 years of disposal. Recycling gives a further reduction of radio-toxicity up to 10 000 to 50 000 years of disposal.
- The long-term residual risk of directly disposed fuel in a thorium matrix is still not known very well, but there are indications on improved performance. Further experimental work is needed to clarify this point.
- Th-assisted Pu burning, using a Th/Pu-MOX type of fuel in a PWR, is an attractive option with respect to mass reduction of Pu.
- Fast neutron reactors and accelerator-driven systems offer both (with similar characteristics) the possibility of a closed Th cycle without make-up fuel, except to start the cycle, reducing mining needs and radiological risks. Full recycling of actinides gives impressively low radio-toxicity results for the wastes over a long period of disposal, starting after the bulk of fission products has decayed.

Non-proliferation concerns are also treated in the report.

The interest of the Th cycle should justify a number of experimental developments, in particular:

- Reprocessing and fabrication techniques of Th fuels could be extended from laboratory scale to industrial scale and further optimised.
- Co-extraction of actinides by pyrochemistry in molten salts, aiming at losses of the order of 0.1% should be demonstrated.
- Nuclear data and physico-chemical data should be established or improved.
- Some simple irradiation experiments should be foreseen, (as it is the case in the new thorium project for the EU 5th FWP).

Finally, the considerations of [37], further enhance the potential of thorium, if powerful accelerators are used.

5.4 Multipurpose neutron source installations

Recently, the "transmutation" community has become involved in the discussions around "multipurpose" facilities, based on a high power proton accelerator, which provides neutrons, by spallation on one (or several) target(s) for different applications.

The most known example is of course the joint KEK-JAERI project [19]. A new initiative is under study in Europe. The ADS experimental installation could be one of the "potential" customers of the neutrons (as it is in Japan), and, consequently, the transmutation community could be interested both to the possibility to demonstrate the concept, and to irradiate the dedicated fuels and targets needed to assess feasibility (see §4).

However, the need to single out a "leading" customer, can somewhat jeopardise the performance allowable for the "lesser" customers. A typical example, is the debate on the pulsed or continuous mode of operation of the high intensity proton accelerator.

6. Healthy criticism

In the reference quoted at the beginning of this report, Rasmussen and Pigford express their doubts about the value of P&T with arguments that should be carefully taken into account still today. Three of their arguments seem of particular relevance, besides the economical and institutional issues, which, although of fundamental importance, have to be adapted to each specific situation:

1. The total inventory of untransmuted radioactivity in the reactor and fuel cycle must also (besides what is sent to the repository as losses at reprocessing) be considered as a potential waste and it takes centuries to reduce it.
2. In the search for an adequate measure of performance, the repository "intrusion" scenario, is claimed to be the most affected by P&T. However, if one considers "intrusion" in a repository, why not to consider "intrusion" in the installations of the fuel cycle, where most of the inventory is kept!
3. P&T will increase to significant amounts new secondary wastes.

As far as arguments 1 and 2, it is clear (and should be always made clear in front of any type of audience), that P&T strategies are definitely associated to an (expanded) use of nuclear energy, with fuel (re)processing and relevant investments in new facilities. However, this (expanded) use of nuclear energy can be made acceptable to the public by the very fact that the burden to repositories is reduced by P&T, and that potentially physical means to eliminate all nuclear materials are provided by the same P&T technologies, even if they should be operated for long periods of time.

Finally, the problem of secondary wastes, and the more general problem of the impact of P&T on the fuel cycle installations, often mentioned in the present report has to be carefully quantified, in particular in terms of social acceptability.

In conclusions, arguments against P&T can be seen simply as arguments in favour of a simplified fuel cycle, and not necessarily in favour of the once-through cycle based on Uranium utilisation.

7. Conclusions and perspectives

Transmutation of wastes has been revisited in the last decade and, although no spectacular breakthrough has been made, a number of significant results have been obtained.

Besides the relevant results in the aqueous chemical separation process domain (which have not been reviewed here), one can quote:

- Understanding of the physics of transmutation and of the "neutron availability" concept.
- Understanding of the role of innovative fuels (including molten salts and particle fuels) to improve the characteristics of the fuel cycle and to minimise wastes.
- Understanding, in that context, of the potential of pyrochemical processes both for fuel fabrication and for irradiated fuel reprocessing.

- Understanding of the role of ADS to handle Pu and MA, but also to provide an option for an extended use of the thorium cycle.
- Understanding of the role of fast neutron spectra and their flexibility. In this frame, the discussion around the coolants for FR would benefit from a better international agreement on pro and cons of the different options.

Since fuels play a central role in all scenarios of waste minimisation and nuclear power development, an international share of efforts around nitrides, oxides and metals should be organised in order to insure an optimum use of resources in the few existing laboratories to handle very active fuels. In that frame, the availability of irradiation facilities, in particular able to provide fast spectra (and high damage rates) is a key point and a major concern.

No convincing case can be made in favour of transmutation, without the full experimental demonstration of its feasibility. Experiments are then needed and the relevant installations should be kept available, with enough experienced teams. Besides the case of the installations for fuel characterisation, fabrication and irradiation, often mentioned in this report, installations related to basic physics (nuclear data and neutronics) will remain vital for all scenarios of development.

In the field of ADS, the development of high power proton accelerators and the construction of a $60 \div 100$ MW_{th} Experimental facility, at a realistic but not too far away, time horizon, seem to be necessary in order not to loose credibility.

International initiatives should be upgraded and, besides the very valuable information exchange goal should address the practical share of work in key fields and should help to focus on some most promising concepts, promoting joint experiments and avoiding dispersion of efforts. In this respect, a co-ordinated activity on pyrochemical processing is strongly suggested.

Finally, the results of the new study of OECD/NEA presently underway will certainly help to better understand and to agree on the relative merits of two of the major options (i.e. critical fast reactors and ADS) for waste transmutation [39].

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