ADVANCED FUEL CYCLES FOR ACCELERATOR-DRIVEN SYSTEMS: FUEL FABRICATION AND REPROCESSING

Institute for Transuranium Elements

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EXECUTIVE SUMMARY

For the fuel cycle of accelerator-driven systems so-called advanced fuels are envisaged. These advanced fuels are characterised by a high content of plutonium and minor actinides, are preferably uranium-free and will be irradiated to relatively high burnup to achieve a high extent of transmutation. In order to select the appropriate fuel material (metal, oxide, nitride, etc.) and the fuel form (pellet, particle) the effects of the choice of the reactor coolant (gas or liquid metal) and, in case of a multiple recycle strategy, the possibility of reprocessing must be taken into account.

Fuel

The choice for the advanced fuel is not obvious at present. Oxides and nitrides are considered the most promising fuel materials, metals and carbides have been discarded. Oxide phases (mixed transuranium oxide, inert matrix mixed oxide, or thorium-based mixed oxide) have the advantage of high chemical stability and thus relative simple handling and fabrication, which is very important for MA-containing materials. The relatively low thermal conductivity of oxide materials, however, will lead to a high operating temperature. Composites (ceramic-metal or ceramic-ceramic) may help to improve this situation. Nitride fuels, in contrast, has much better thermal properties and, hence, a low(er) operating temperature but are more difficult to fabricate. In addition, the relatively low chemical stability of the transuranium nitrides may lead to severe safety problems during power/temperature excursions (nitrogen pressure build-up, actinide metal vaporisation/- redistribution). A general problem for fuel with high MA content is the helium produced by transmutation. To deal with this, the standard pellet form may need modification or other fuel forms, e.g. particles, may be considered.

The primary or reference option for the advanced fuels for ADS is based on the $(Pu,MA,Zr)O_2$ or $(Pu,MA,Th)O_2$ material: either as a homogeneous phase or a composite in which it is distributed steel (CERMET) or MgO (CERCER). An optimisation of this fuel type needs to be performed in the coming years in a European research and development programme, during which a selection of the fuel form (standard pellets, modified pellet, or particle) will be made. The back-up option for the advanced fuel is (Pu,MA,Zr)N.

Fuel processing

Two types of processes can be applied to the separation of long-lived radionuclides: hydrochemical ("wet") and pyrochemical ("dry") processes. The PUREX process is the industrial hydrochemical reprocessing technique to separate U and Pu from spent fuel and is based on the dissolution of the fuel in nitric acid. For the extraction of minor actinides the process should be modified/extended for which extensive research is being made at present. Neptunium could be extracted in a modified PUREX process, americium and curium in the extended PUREX process in which additional extraction steps follow the standard process. This extension must include the separation of minor actinides from the lanthanides, which are generally co-extracted due to

very similar chemical properties. An alternative to hydrochemical methods are pyrochemical processes in which refining is carried out in the absence of water in a molten salt. In nuclear technology they are often based on electrorefining or on distribution between non-miscible molten salt-metal phases.

The major advantages of pyrochemistry to reprocess advanced fuels, in comparison to hydrochemical techniques, are a higher compactness of equipment and the possibility to form an integrated system between the irradiation and reprocessing facilities, thus reducing considerably transport of nuclear materials. Especially for advanced oxide fuel (mixed transuranium, inert matrix or composite) and metal fuels, but also nitride and carbide fuels, pyrochemistry is to be preferred. In addition, the radiation stability of the salt in the pyrochemical process compared to the organic solvent in the hydrochemical process offers an important advantage when dealing with highly active spent MA fuel. This will allow shorter cooling times which reduce storage costs.

Because hydrochemical reprocessing of the proposed advanced oxide fuels is difficult, it is clear that pyrochemical reprocessing techniques must be considered. This means that direct electrolysis of the oxide must be developed. Alternatively a conversion process could be envisaged to convert the oxide to a metal or to a chloride that can be dissolved in the molten salt.

Infrastructure

In Europe facilities for the fabrication of americium and curium fuel for R&D purposes exist at the Institute for Transuranium Elements (JRC, Karlsruhe) and in ATALANTE (CEA, Marcoule). In these facilities fuel pins for characterisation and the first irradiation tests can be made. The capacity is limited to a few grams of curium and tens of grams for americium. For larger scale irradiation tests a (small) dedicated fabrication facility needs to be constructed.

The first irradiation tests of these advanced fuels can be performed in existing irradiation facilities. In Europe this will mainly be Materials Testing Reactors, in which spectrum tailoring can help to achieve a representative neutron spectrum. In the longer term, however, tests in fast systems are unavoidable. These tests should be performed in the European experimental ADS facility (XADS). Post-irradiation examination of these fuels can be performed in existing hot-cell facilities.

1. INTRODUCTION

Transmutation of long-lived radionuclides using accelerator-driven systems (ADS) is attracting much attention worldwide. In an ADS a high-power proton accelerator is used to produce neutrons in a heavy-metal target (W or Pb) which are used to "transmute" transuranium elements and long-lived fission products in a subcritical reactor core. The transuranium elements are "transmuted" by fission reactions, thus producing additional neutrons and energy. Some long-lived fission products are "transmuted" by neutron capture converting them into short-lived or stable radionuclides.

In order to demonstrate the potentiality of the ADS concept, research programmes are presently being defined in Europe [98Rub], USA [99DOE], and Japan [97Muk]. In Europe a common platform on ADS has been formed among several member states of the European Union with the goal to establish a partnership between research agencies and at the industrial level. A Technical Working Group (TWG) has the task to review the ongoing actions in the participating countries and to aid the convergence towards a common European initiative for a demonstration plant [98Rub]. The TWG has issued an interim report on the feasibility of a 100 MW ADS demonstration facility for Europe, the so-called XADS, to be built in 2010-2015. Two concepts for the ADS are considered: gas-cooled and liquid metal cooled (lead or lead/bismuth). The TWG has also issued a report on the status of relevant R&D in the EU countries as well as the R&D proposals related to ADS for the 5th Framework Programme.

Several technical issues that are considered critical in the development of ADS are to be addressed by the TWG. A special expert group, the Fuel Fabrication and Processing subgroup, has been established to deal with the fuel cycle issues. The mandate of this subgroup (see Appendix A) covers two technical areas:

1. The fuel for the X-ADS.
2. Advanced fuel cycles for future ADS transmutation devices.

The latter will be discussed in detail in the present report.

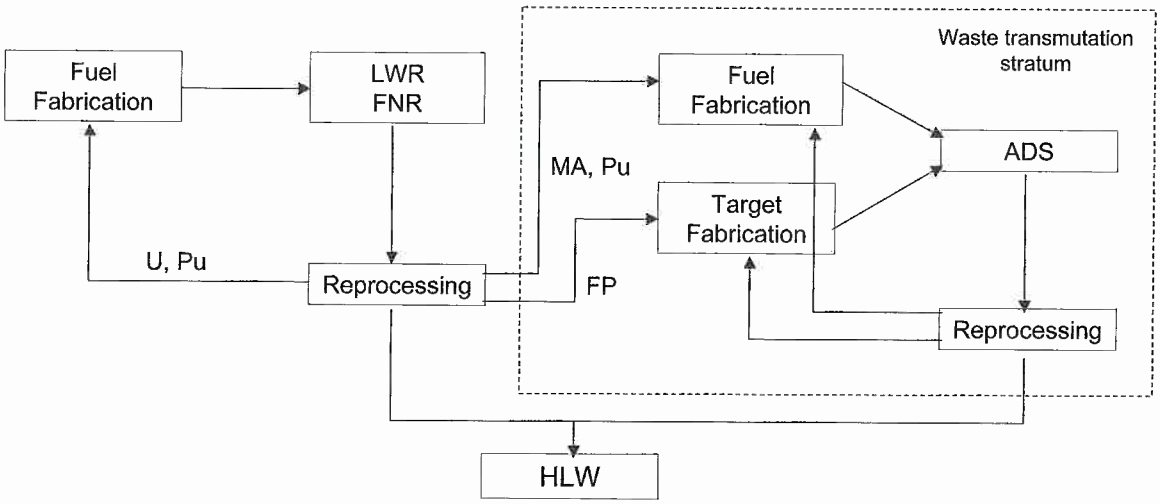

Figure 1.1: Schematic presentation of the dual strata fuel cycle.

Fuel cycles for ADS transmutation devices are generally described as "dual strata" as there is a distinct difference between the conventional "energy production stratum" consisting of LWRs (and possibly FNRs in the future), and the "waste transmutation stratum" in which dedicated technologies are being employed (Figure 1.1). The present report is restricted to the second stratum: the state-of-the-art for various fuel options and reprocessing options (hydrochemical and pyrochemical) are described and the key areas for research are identified. However, since the reprocessing in the first "stratum" forms the link with the second stratum, some attention will also be given to the possible improvements of the conventional PUREX reprocessing. In addition, an evaluation of the availability of the required research facilities is presented. Based on this information, recommendations for the fuel cycle of an European ADS transmutation device are made.

2. ACTINIDE FUELS AND TARGETS

2.1 General considerations

Many different types of fuels or targets for transmutation of actinides have been suggested in the recent years, mainly depending on fuel cycle strategy considerations. Two general approaches can be distinguished in this context:

i. The mixing of small quantities of the minor actinides (up to about 2.5 w%) together with the plutonium to the fuels of standard cores. This has, however, a big impact on the fuel cycle because the number of MA-containing subassemblies would be relatively high and the effect on the core reactivity coefficients would be considerable.

ii. The use of dedicated fuels/targets, which have a high content of plutonium and minor actinides, in special actinide "burners", like an ADS. The number of subassemblies required for the same mass throughput is much less than that for (i). In addition, the impact on the core reactivity can be reduced by optimising the design of the subassemblies.

In the present report only the latter approach is considered. We will principally focus on material containing only plutonium and minor actinides as fissile/fertile material in a ratio that may vary between 1:5 to 5:1. Such material is considered as a fuel when the isotopic composition gives rise to a critical or a reasonably subcritical core, and a target when it does not contribute significantly to the criticality of the reactor core.

It is obvious that when a fuel or target does not contain ^{238}U, the transmutation efficiency is higher because no new actinides are formed. In that case a non-fissile (inert) matrix may be considered as support or diluent of the actinide phase. In a homogeneous fuel or target, the diluent forms one phase (a solid solution) with the actinides, in a heterogeneous fuel or target the actinide phase coexist with the diluent (a dispersion). Of course, combinations of the two concepts or non-diluted fuels or targets need to be considered as well.

As a result of the high content of plutonium and minor actinides, the radioactivity of the virgin fuel material is much higher than in case of UO_2 or MOX fuel, and special protection is required during the fabrication step to shield gamma and neutron radiation. This will lead to special requirements for the fabrication processes, such as remote handling or, preferably, automated processes, which need to be considered as a criterion. This, of course, will also apply to subsequent transport and intermediate storage.

Furthermore, a high burn-up of the fuel/target is required to achieve a reasonable extent of conversion in the transmutation process. This will lead to high demands on the performance of the fuel in view of the concomitant high fission-gas production, fuel swelling and fuel cladding chemical and mechanical interaction, which will exceed the levels known from existing fast reactor fuels.

Table 2.1: Most important characteristics of dedicated fuels, divided in intrinsic and extrinsic (shaded) factors.

Characteristic		Effect	Criteria for fuel and fuel pin
1	High Pu+MA content	Multi-element composition	Solubility/compatibility of the phases
		High linear rating	Good thermal conductivity of fuel pellets
			High melting point fuel material
		Helium gas production	Low smeared density
		Volatility of fuel components	Low fabrication and operation temperatures
		High gamma and neutron dose	Shielded fabrication
2	U-free	Pu+MA only	Design optimisation
		Inert support	Compatibility of support and actinide phase
3	High burn-up	High fission gas production	Low smeared density
		Fuel swelling	Low smeared density
		Fuel cladding interaction	Advanced cladding material
		High level of radiation damage	Radiation stability fuel material
4	Pb(/Bi)- cooled	Possible interaction	Compatibility of cladding with molten lead
			Compatibility of fuel with molten lead
5	Gas-cooled	High(er) cladding temperature	Stability of cladding material
		High(er) fuel temperature	Good thermal conductivity of fuel pellets
			High melting point fuel material
6	Reprocessing	Aqueous processing	Dissolution in HNO_3
		Pyrochemical processing	Direct electrolysis or conversion
7	Accelerator-driven	Regular beam trips	Thermal shock resistance

These and some other general criteria for dedicated fuels and targets that will be considered are listed in Table 2.1. In addition to these, reactor physical and safety characteristics need to be considered, but this will need quantitative analysis that is outside the scope and the mandate of this group.

2.2 Metal fuel

2.2.1 Characteristics

The actinide metals have a complex allotropy as a result of which alloys of pure actinides undergo several phase transitions in the temperature range of operation of the fuel. In metal fuel, the cubic gamma phase of the alloy should be stable at the operating conditions. The mutual solubility of the actinide metals is not well known. The limited information available shows that Np and Am are not miscible in all proportions [93Oga]. As a result it is difficult to obtain a homogeneous alloy if the Am and Np contents are significant.

The thermal conductivity of metallic fuel is high. For uranium-based alloys, for which the melting temperature is around 1623 K (1350 °C), this gives a reasonable margin to melting. However, for U-free and Pu-rich alloys, the situation is different, as the melting point of Pu (913 K) and the thermal conductivity of Pu metal are significant lower than that of U (melting point 1408 K). Moreover, americium metal has a relatively high vapour pressure, which requires a lower operating temperature to avoid actinide relocation. Adding a non-fissile metal with a high(er) melting point and a high(er) thermal conductivity is thus a requirement to increase the melting point of the alloy and raise the margin to melting. Candidates are molybdenum (2896 K), zirconium (2128 K), titanium (1941 K), or niobium (2750 K). Zirconium is the most promising of these and a lot of experience already exists in the USA with addition of Zr to U-Pu metallic fuels. However, the amount of "matrix" that can be added to the alloy is subject to phase considerations and technological restraints from the fabrication. Moreover, Np and the low temperature alpha form of Zr are only partially miscible in the whole composition range [94Jul, 94Gib]. Intermetallic compounds can be formed, which could precipitate in the metallic alloy and could dissociate in Zr and Np at low temperature (550°C). In contrast, Np is very soluble in the U-Zr alloy [94Jul]. As a result local melting of Np in U-free alloy at low temperature (640°C) cannot be excluded when the Np content in the fuel is significant. In the US ATW concept it is therefore proposed to mix the Pu-MA-Zr alloy in a Zr metal matrix (see section 2.6).

A second option to keep sufficient margin to fuel melting generally applied in metallic fuel designs, is the use of a metallic bond (liquid sodium) between pellet and cladding to enhance heat conduction from the fuel to the coolant [98Wal]. This is necessary whatever the operating conditions.

U-Pu metal fuels shows significant swelling at relatively low burn-up as a result of anisotropic growth. Addition of zirconium to the alloy can reduce this. With increasing burn-up, also the accumulation of fission gases and, in case of minor actinides, helium in bubbles contributes to the swelling. At a fuel swelling of about 30%, the gas bubbles in the fuel start to interconnect to form paths for release. Therefore the swelling can be accommodated by increasing the space between the fuel and the cladding (lowering the smeared density) and increasing the plenum. This is possible because of the excellent thermal properties of the metallic fuel and the possibility of applying metal bonding. In metal-fuelled fast reactors such as EBR-II, the metal bond and the coolant are identical (sodium). When liquid lead or lead/bismuth are considered for ADS, this is not possible as plutonium and zirconium are not compatible with Pb or Bi. They form intermetallic compounds or partially dissolve in the molten metal.

2.2.2 Fabrication

The most common fabrication process for metallic fuel is injection casting of the alloy. In this process the alloy is heated in an evacuated furnace to a temperature above the melting point and cast into (quartz) moulds, in which the alloy solidifies quickly. This process has been used to fabricate the U-Pu and U-Pu-Zr fuels for the EBR-II in the USA [97Hof]. This fuel was not completely homogeneous but consisted of both alpha- and beta-uranium phases. The injection casting method is very suited for remote handling and automation. However, at the elevated temperatures needed in this process, some problems may be expected in case of americium-containing alloys since Am metal is relatively volatile and may evaporate. To recover and re-use this material, a trapping system must be designed.

Powder metallurgy (mixing, pellet production, sintering) is therefore also being considered. For example in the US ATW proposal powder metallurgy is proposed to produce the Pu-MA-Zr alloy [99DOE]. The alloy, which can be obtained directly from pyrochemical processing, is processed into powder of small particle size. Alternatively, the alloy can be produced by co-melting of the components, but in that case the vaporisation of americium has to be avoided. However, this method requires technological safety measures to due to pyrophoric nature of the actinide metal, enhanced by the high surface area of metal powders.

2.2.3 Irradiation behaviour

Experience from (U,Pu) fuel

Irradiation data for metal fuels are only available for U-Pu fuels, in particular from for U-based metal fuels from the fast breeder programme in the USA and the evolving IFR concept of Argonne National Laboratory. Considerable experience has been gained with uranium metal fuel that has been used as driver fuel in the EBR-II [97Hof]. Up to 30.000 fuel pins have been irradiated regularly up to a burn-up of 8 at% and safety tests on irradiated fuel were performed. Also some U-Pu-Zr fuel pins have been tested successfully in the EBR-II reactor up to 20 at% burn-up. It was found that the steady-state performance was good. During off-normal conditions, fuel expansion was observed and at high temperatures a reaction between fuel and the stainless steel cladding occurred (interdiffusion). An irradiation test of (U,Pu,Zr) with up to 5% minor actinides in the Phénix reactor is being planned by the Japanese research institute CRIEPI. The metallic fuel for this experiment was fabricated by casting at ITU [97Kur].

TRU fuel

Data on uranium-free alloys are not available.

2.2.4 Summary

Metal fuel has interesting thermal and mechanical properties, which can be improved further by sodium bonding. Metal fuel based on U could be an interesting option but does not meet the criteria of this study. A mixed transuranium metal fuel does not seem feasible (low melting temperature, limited mutual solubility). Addition of a non-fissile metal in a significant quantity is required. Zirconium seems to be the most likely candidate but there is considerable uncertainty about the (mutual) solubility of neptunium and zirconium. For the fabrication process of such fuel considerable technological measures need to be implemented due to the pyrophoric nature of metals and the high volatility of Am and Pu. In addition, the compatibility of metal fuel with liquid lead or lead/bismuth is not certain. Also significant Am redistribution will probably occur during irradiation.

2.3 Oxide fuels

2.3.1 Mixed transuranium oxide fuel (homogeneous/undiluted)

2.3.1.1 Characteristics

The transuranium dioxides PuO_2, NpO_2, AmO_2 all have a fluorite-type crystal structure and their lattice parameters are very close. As a result the $(Pu,Np,Am)O_2$ solid solution can probably be prepared in a wide range of compositions. CmO_2 is also cubic, but this compound is thermodynamically unstable with respect to Cm_2O_3: the decomposition temperature is about 650 K. In a $(Pu,Np,Am,Cm)O_{2-x}$ solid solution, the curium is therefore present as Cm(III).

Compared to standard MOX fuel, significant differences in the physico-chemical properties can be expected for the $(Pu,Np,Am,Cm)O_{2-x}$ solid solution (mixed transuranium oxide, MIMOX):

1. Lower melting point. The melting point of the actinide dioxides decreases continuously from UO_2 to AmO_2 (see Table 2.2). The $(Pu,Np,Am,Cm)O_{2-x}$ solid solution thus will have a significantly lower melting point, and smaller margins to melting compared to conventional (U,Pu)-mixed oxide fuel.
2. Higher oxygen potential of AmO_2 due to its lower thermodynamic stability. Depending on the fuel composition and its oxidation state, this might lead to more severe cladding corrosion, especially at high burn-up.
3. Higher volatility of Am species and, to a smaller extent, of Pu species. This could result in difficulties during the fabrication of the fuel pellets and to actinide redistribution during the irradiation due to temperature gradients in the fuel.
4. Lower thermal conductivity. Uncertainties still exist about the thermal conductivity of some of the minor actinide oxides. It seems that the thermal conductivity decreases from NpO_2 to AmO_2, though the experimental values reported in literature for the latter compound [86Sch]

are highly uncertain [98Bak]. New measurements are needed. However, measurements of the thermal conductivity of $(U,Pu)O_2$, $(U,Np)O_2$, $(U,Am)O_2$ and $(U,Np,Am)O_2$ made in the frame of the SUPERFACT experiment, show that addition of minor actinides to UO_2 leads to some decrease of the thermal conductivity, although the changes are only significant in the case of $(U,Am)O_2$. Anyway, the lower thermal conductivity would lead to a higher fuel temperature compared to UO_2 or MOX fuel and thus to a smaller margin to fuel melting and a higher fission gas release.

Table 2.2: Some properties of the actinide dioxides

		ThO_2	UO_2	NpO_2	PuO_2	AmO_2	CmO_2
Structure [a]		fcc	fcc	fcc	fcc	fcc	fcc
Melting point/K		3640	3113	2823	2663	2448	Decomp.
Thermal conductivity [b] / W m^{-1} K^{-1}	1273 K	3.0	3.1	?	3.0	?	-
	2273 K	1.9	2.7	?	2.7	?	-

[a] fcc = face centered cubic

[b] corrected to 100% theoretical density

2.3.1.2 Fabrication

There is a large know-how of design and fabrication of oxide fuel since more than 40 years. The current oxide technology is based on powder metallurgy. The actinide oxide powders are homogenised, compacted and sintered to form fuel pellets. This technology has been applied on Am-containing materials at laboratory scale. The main drawback of such a process is that some fabrication steps are not easily adaptable to remote handling in shielded cells as is required for Am- and Cm-containing fuels. Furthermore the handling of powders results in the production of radioactive dust that accumulates in the glove boxes, leading to increasing dose rates. Therefore fabrication of such compounds will require a significant R&D effort to simplify as much as possible the process and the tools, from the powder synthesis to the clad filling. For example, powder mixing can be avoided by the production of a mixed powder at the reprocessing stage, for example by sol-gel coprecipitation of Pu+MA from the nitrate solution. The sol-gel route has been found suitable for MA fuels in the SUPERFACT experiment. Also the potential of non-pellet fuels, such as (low density) sphere-pac or crushed particles packed by vibration (e.g. the Russian VIPAC process) must be investigated as a potential fabrication route for dedicated fuels because compaction and, in some processes, sintering can be avoided.

2.3.1.3 Irradiation behaviour

Experience from (U,Pu) fuel

Significant irradiation experience exists for uranium-based mixed-oxide fuel containing minor actinides (MINOX). In the SUPERFACT experiment fuel pins containing various quantities of minor

actinides (Np,Am) were irradiated in the Phénix reactor [96Cha]. The main purposes of the experiment were the analysis of the general irradiation behaviour of Minor Actinides containing fuels and the comparison between fuels with high MA and low MA content. Eight fuel pins of four different types of fuels were prepared:

- 2 pins with $(U,Pu)O_2$ and 2% NpO_2
- 2 pins with $(U,Pu)O_2$ and 2% AmO_2
- 2 pins with UO_2 and 45% NpO_2
- 2 pins with UO_2 and 20%NpO_2 and 20% AmO_2

The pins were irradiated (together with "standard" pins of Phénix) for 4 cycles, resulting in approximately 4.5 at% burn-up for the pins with high MA content and approximately 6.5 at% burn-up for the pins with low MA content. After PIE the main results were as follows:

- Good behaviour of all fuel variants similar to "standard" MOX-fuel of Phénix.
- Beginning of mechanical interaction between fuel and cladding for the pins with high MA content.
- The depth of corrosion was as expected (maximum 60μm).
- The fission gas production and release was as predicted for the given power.
- A higher He-production in the presence of Am, mainly due to daughter products with high specific alpha activity (^{242}Cm and ^{238}Pu). The released He contributed to an increase of the internal pin pressure.
- The extent of transmutation of Np and Am were measured after irradiation and for both elements a mean value of about 30% was found

In the experiment TRABANT1 two mixed-oxide fuel pins were irradiated in a "TRIOX"-capsule of the HFR-reactor. The purpose of the experiment, which was launched in the frame of the CAPRA-programme, was the study of the behaviour of mixed oxide fuels with a very high Pu-content (pin 1), with a high Pu-content and an addition of neptunium oxide (pin 2). The fuel compositions were $(U_{0.55}/Pu_{0.45})O_2$ for pin 1 and $(U_{0.55}/Pu_{0.40}/Np_{0.05})O_2$ for pin 2. Pin 1 was fabricated by mechanical powder mixing at CEA, pin 2 was fabricated by a SOL-GEL precipitation procedure at ITU. The pins were irradiated to approximately 10% burn-up. PIE showed, that pin 1 had failed during irradiation (around 5-6% burn-up) most likely due to an "early corrosion attack" (corrosion de jeunesse), i.e. it is assumed that the linear rating of the pin at beginning of the irradiation was too high, producing a high amount of aggressive gaseous fission products [2000Pla]. The examination of pin 2 (which is under progress now) did not show any irregularity and no sign of a defect. It should be noted that this pin operated at a somewhat lower linear power.

TRU fuel

Irradiation data for mixed transuranium oxide fuels are not available. The existing information is restricted to preliminary calculations of the performance of $(Pu_{0.5},Am_{0.5})O_2$ fuel pin of CAPRA type (fuel temperature and actinide redistribution versus burn-up) performed in the frame of the CADRA programme [99Pil], taking into account the available data on actinide oxides. The results showed that the O/M ratio of the fuel and the oxygen potential in the $(Pu,Am)O_2$ fuel lead to lower partial pressures of the actinide-containing gases and hence the redistribution of Am and Pu is less

significant than expected. However, many basic data are missing for $(Pu,Am)O_2$ and they were assessed theoretically (melting point, oxygen potential and chemical behaviour of Am) or extrapolated from MOX fuel (in particular thermal conductivity, swelling and gas release models). Clearly these results have to be confirmed experimentally. The calculations showed the technical feasibility of homogeneous mixed transuranium oxide pins, provided the design is optimised to guarantee sufficient margin to the melting and to accommodate the expected swelling due to the helium production. Small diameter pins or annular pins with inner cooling could be attractive solutions which deserve to be analysed in-depth in case of Pb or Pb/Bi cooling. For gas cooled ADSs, the uranium-free mixed-oxide pin concept is probably less attractive because it can not ensure sufficient margins to melting, at least in case of gas depressurisation. However, other interesting solutions for mixed transuranium oxide compounds remain like inert matrix fuel pins or particle fuels (see sections 2.6 and 2.7).

2.3.2 Inert-matrix mixed-oxide fuel (homogeneous/diluted)

2.3.2.1 Characteristics

A limited number of compounds are considered as a host for the actinide oxides in so-called inert-matrix mixed oxide fuel (IMMOX), such as ZrO_2, CeO_2 and Y_2O_3. A large development programme is currently being performed on stabilised zirconium oxide for plutonium burning in LWRs [2001Led]. The attractive properties of this material are the small neutron capture cross sections of the components, the high melting point, the high radiation stability and the high chemical stability. Yttria-stabilised (YSZ) as well as calcia-stabilised zirconium oxide (CSZ) are considered because with the addition of dopants such as CaO or Y_2O_3 the cubic fluorite lattice of ZrO_2 can be partially or completely stabilised down to room temperature. In addition, actinide oxides can act as stabilisers of the cubic structure. The substitution with aliovalent ions (e.g. tri- and bivalent, but not tetravalent) in zirconium oxide also creates anion vacancies in the crystal structure. The structure of the yttria-stabilised zirconium oxide is similar to that of the actinide dioxides, and the solubility of PuO_2 and AmO_2 is therefore high. The phase diagrams of the systems Zr-Y-An-O are not very well known, especially in case of the minor actinides. The existing information about the ZrO_2-PuO_2 binary indicates that the melting point of the solid solution is close to that of PuO_2. However, in case significant quantities of americium are added a significantly lower melting point can be expected.

The thermal conductivity of stabilised zirconium oxide is rather low. This will lead to a high central temperature of the fuel and limit the margin to melting. The temperature dependence of the thermal conductivity of zirconia-based inert-matrix fuel was studied by Pouchon [2000Pou] who suggested a conductivity value of 1.9 $W \cdot m^{-1} \cdot K^{-1}$ for the simulated fuel $(Zr_{0.75}Y_{0.10}Ce_{0.10}Er_{0.05})O_{1.925}$, independent of the temperature. The effect of plutonium is not yet know but experimental measurements are under way [2001Deg].

In addition to zirconium oxide, cerium oxide has been suggested as matrix for IMMOX fuels. The dioxide CeO_2 has a cubic structure like the actinide dioxides and thus can form a solid solution. The thermal conductivity of CeO_2 is even lower (1.2 $W \cdot m^{-1} \cdot K^{-1}$) than that of YZS and is very dependent on the O/M ratio of the oxide. Furthermore, the relatively high oxygen potential will very likely lead to cladding oxidation.

2.3.2.2 Fabrication

IMMOX fuels can be prepared by powder mixing or by wet chemical routes such as co-precipitation or sol-gel processes. The aqueous processes are to be preferred in case of minor actinide containing fuels as they avoid the mixing and milling of powders, which is an unwanted fabrication step because of the dust formation. However, the chemical stability of the solutions containing significant amount of transuranium elements (producing hydrogen gas as a result of radiation-induced dissociation of water) has to be evaluated.

For the aqueous processes nitrate solutions containing the elements composing the final product are used. In case of co-precipitation, ammonia is added under strong stirring to obtain a precipitate (powder) that can be used for compaction into pellets. In case of the sol-gel processes there are two possibilities, internal gelation and external gelation. The principle of these processes is the conversion of the nitrate solutions to oxides, based on the co-precipitation of metal hydroxides by the addition of ammonium hydroxide. In both cases spherical particles are obtained, which are washed, dried, calcined and sintered resulting in a dense oxide product. Depending on the technique monodisperse or polydisperse particles can be obtained.

The co-precipitation route and internal gelation have been shown to be successfully working for Pu- and non-Pu-containing YSZ-fuel by PSI [96Led, 2001Led]. The external gelation process is being tested in several other laboratories (ITU, NRG, CEA). An alternative method for the fabrication of YSZ-based actinide fuels, based on the external gelation method, is being developed at ITU [97Ric]. In this process the actinides are infiltrated into spherical particles produced by a sol-gel technique. The sphere production can be performed outside the nuclear environment and only the relatively simple infiltration step, which can be automated easily, needs to be performed in the glove box. This process has been demonstrated on a laboratory scale with Ce and, recently with Pu [2000Fer].

2.3.2.3 Irradiation behaviour

The data on the irradiation behaviour of transuranium homogenous inert matrix fuels are very limited. In the 1960s irradiation tests of $(Zr,Pu)O_2$ fuel at high linear heating rates were performed in the ETR reactor for two compositions (1.93 and 9.76 wt% PuO_2) [63Han]. The fuel with the highest Pu content, which was operated close to the melting point, developed a central hole

and columnar grains. It was found that in these samples the fission products and PuO_2 have migrated. The results of this investigation have demonstrated the satisfactory irradiation performance of this type of fuel.

In the experiment TRABANT1 two half-columns of the $(Pu,Ce)O_2$ fuel were prepared (pin 3), which differed in the oxygen/ metal ratio (2.00 and 1.96 respectively). The pin was irradiated (together with the mixed oxide pins of TRABANT 1) at a higher linear power than initially planned. In the first intermediate examination by neutron radiography it was recognised that the half column with the fuel with the low O/M ratio showed melting phenomena. Theoretical calculations showed that melting is explained by the very low thermal conductivity of the substoichiometric fuel type which is not compatible with the high operating power.

An irradiation test of $(Zr,Y,Er,Pu)O_{2-x}$ fuels (fabricated by PSI) started at the OECD Halden reactor (thermal spectrum) in 2000 (IFA-651). After a pre-conditioning at 235 W/cm during the first 100 days of irradiation; the fuels will be irradiated for 900 days at a peak linear power rate of 350-400 W/cm. This means that a rather high burn-up will be reached. Bulk pellets are tested, whereas thermocouples are inserted in annular pellets. The pins are equipped with thermocouples, pressure and stack elongation sensors in order to have information about fuel behaviour and fission gas release. The same material fabicated by PSI is being irradiated in an irradiation experiment on the incineration of Plutonium using inert matrix fuels that is ongoing in the HFR-Petten [2001Sch]. In this experiment also a U-doped $(Zr,Y,U,Pu)O_{2-x}$ fuel sample (JAERI concept) is included, as well as a MOX sample for reference.

2.3.3 Thorium-based mixed-oxide fuel (homogeneous/diluted)

2.3.3.1 Characteristics

Although thorium oxide is fertile (it produces ^{233}U upon irradiation), the production of plutonium and minor actinides in thorium-based fuels or targets is limited. The properties of ThO_2 as a matrix phase are very promising: it has a cubic fcc structure like the other actinide dioxides, it has a high melting point and a thermal conductivity close to that of UO_2. Moreover, ThO_2 is very radiation resistant and thus allows reaching high burn-up.

2.3.3.2 Fabrication

The fabrication of thorium oxide pellets, spheres, or coated particles is well known. $(Th,Pu)O_2$ pellet fuels have been produced in the past in USA and India. In Europe the fabrication has recently been resumed for irradiation experiments in the frame of the 5th European Framework Programme. Standard powder blending (comparable to MOX) is well suited for this fuel but also wet chemical routes such as co-precipitation or sol-gel processes can be used. It needs to be mentioned that the

sol-gel technique was demonstrated extensively for the production of microspheres for the HTGR fuel.

Of course these fabrication routes need to be extended to $(Th,MA)O_2$ taking into account the increased radiation dose, as discussed in previous section. The experience from the SUPERFACT experiment has shown that the sol-gel route is very well suited for the production of $(U,Pu,MA)O_2$ and it can be expected that this is also true for $(Th,Pu,MA)O_2$.

2.3.3.3 Irradiation behaviour

Experience from (Th,U) fuel

There are not many irradiation data available for thoria fuel; only a limited number of experimental fuel-test programmes have been performed. The majority of the data comes from the USA where thoria fuels for LWRs have been tested in the past in the LWBR programme. But also in Europe development programmes for LWR thoria fuel were performed. Hart et al. reviewed the irradiation performance of thoria-based fuels [79Har] for LWRs and HWRs and came to the conclusion that $(Th,U)O_2$ is comparable to, and in some respects superior to UO_2. However, it should be pointed out that the majority of the experiments were performed with low UO_2 content (<10%) and low burnup. Also the irradiation performance of HTGR fuel has been tested [92Han,92Nab] and again little difference between thoria-based and urania-based fuel particles has been observed. Thorium-Uranium-fuel is being irradiated in the Halden material test reactor (IFA-652), started in 2000. The material has been fabricated at Kjeller, Norway, following the procedures of ENEA, Italy. The test is instrumented with thermocouples, pressure and stack elongation transducers. The results will be available through the OECD Halden reactor project.

TRU fuel

In the frame of the 5th European Framework Programme irradiations of $(Th,Pu)O_2$ will be started in the HFR-Petten and in Kernkraftwek Obrigheim (Germany), though for compositions and conistions that are specific for LWRs. Results of these experiment will become available in 2003/2004 and thereafter. No irradiation data exist for thorium-based fuels containing minor actinides, but, again, the parallel to the SUPERFACT fuels needs to be mentioned (see section 2.3.1.3). In this experiment $(U,MA)O_2$ have shown a satisfactory behaviour of fuel containing 20 wt% Np plus 20 wt% Am, though the burnup was relatively low.

2.3.4 Summary

Mixed transuranium oxide fuel is a logical extension of the current MOX fuel fabrication technology. However, the handling of the minor actinides requires additional shielding and (more) extensive automation due to the increased gamma and neutron emissions. Based on the available information, it can be expected that mixed transuranium oxide fuel has a higher operating temperature than MOX fuel, which is caused by the relatively low thermal conductivity and the low melting point (and as a consequence a smaller margin to melting and possible Am redistribution). This forms a major disadvantage of this fuel type.

Inert-matrix mixed oxide fuel also has the advantage of relatively simple fabrication and handling, not too much different from the current MOX technology when dry processes are used. Like the mixed transuranium oxide fuel, the margin to melting is expected to be small and, in case of zirconium oxide based materials, the operating temperature high as a result of the poor thermal conductivity. Inert matrix fuels therefore do not seem to be a candidate for fuels with high content of actinides operated at medium to high power.

Thorium-based oxide fuel has the highest potential of the mixed oxide, though the thorium matrix is of course not inert. Its fabrication is relatively straightforward and irradiation behaviour is expected to be good from the point of view of thermal and mechanical behaviour.

Engineering solutions (e.g annular pellets, internal cooling) may help to resolve the disadvantages noted for these oxide fuels. Another option is the mixing of the materials with a matrix which improves the thermal properties. This will be discussed in section 2.6.

2.4 Nitride fuel

2.4.1 Characteristics

Nitride fuels have been investigated in past in the frame of the fast-reactor programmes. The advantages of nitrides are the high actinide density (interesting when breeding is the aim), the compatibility with the PUREX process and the chemical compatibility with water, air and stainless steel cladding materials. A major drawback is, however, the production of ^{14}C through (n,p) reactions on ^{14}N in nitride fuels. To be compatible with the current norms of gaseous ^{14}C release in the reprocessing plants, it is necessary to use nitrogen enriched in the isotope ^{15}N, which raises the cost of the fuel. Studies [2001Wal] have shown that it is essential that the ^{15}N will be recycled at the fabrication stage. Also at the reprocessing stage a significant part of the enriched ^{15}N could be recovered when pyro-chemical reprocessing of the fuel is used [98Osu]. But this is still subject to important R&D studies (in particular in Japan).

PuN, NpN, AmN and CmN all have the same crystal structure (cubic NaCl-type) and their lattice parameters are very close. It is thus expected that the solid solution (Pu, Np, Am, Cm)N exists in a wide composition range. The transuranium nitrides have the following characteristics (see also Table 2.3):

- NpN is the only compound in the Np-N system. It forms a complete series of solid solutions in the NpN-UN and NpN-PuN systems. NpN is stable in water and is dissolvable in HNO_3.
- PuN is the only compound in the Pu-N system and has a small homogeneity range on the Pu side at high temperatures. No reactions between PuN and steels were observed up to 600°C. No reactions with Na were found up to 890°C. Fine PuN powder react easily with oxygen and air. Bulk material oxidises more slowly. PuN reacts in boiling water to PuO_2 and is dissolvable in concentrated HCl and HNO_3.
- AmN is the only compound in the Am-N system. No physical and chemical properties are known, but the vapour pressure of N_2 and Am above AmN is supposed to be high since Am usually vaporises during fabrication process of mixed U and Pu nitride, when Pu is old and contains Am. Also significant lattice swelling due to changes in valence state with temperature, as is observed for CeN [97Sch], could be expected. Finally, the hygroscopic nature of AmN is expected to be like rare-earth mononitrides, which implies that greater care must be taken compared to (U,Pu)N fabrication to maintain the inert atmosphere.

Table 2.3: Some properties of the actinide nitrides.

		UN	NpN	PuN	AmN	CmN
Structure		Fcc	Fcc	Fcc	Fcc	Fcc
Decomposition temperature/K		3053[a]	2948[a]	2843[a]	?	?
An pressure/bar	1773 K	$5{\cdot}10^{-10}$	$5{\cdot}10^{-8}$	$1{\cdot}10^{-6}$	?	?
N_2 pressure/bar	1773 K	$5{\cdot}10^{-9}$	$5{\cdot}10^{-8}$	$2{\cdot}10^{-7}$	?	?
Thermal conductivity[b]	1273 K	25	17	13	?	?
/ W K^{-1} m^{-1}	2273 K	28	≈ 18	15	?	?

[a] under 1 bar N_2

[b] extrapolated to 100% theoretical density

The actinide nitrides have a high thermal conductivity compared to the actinide oxides (up to 5 times higher for UN compared to UO_2) whereas the melting temperatures are similar. This results in a lower central fuel temperature or, alternatively, allows a higher actinide content in the target. In case of normal and off-normal operation, the decomposition of nitrides in metal and nitrogen gas (which leads to the formation of a liquid phase in the fuel) is unlikely even at high linear power [99Mig]. However the decomposition could be a critical issue in case of severe accident (core disruptive accident or CDA). There are two major disadvantages for mixed nitride fuels in a sodium-cooled fast reactor:

- The violent reaction of the mixed nitride with sodium, resulting from the high thermal conductivity of the nitride, leading to an explosive heat exchange between fuel and gaseous sodium (Fuel-Coolant Interaction or FCI).

- The pressurisation of the reactor vessel, which can reach up to 50 bar in the extreme case (caused by thermal dissociation of the nitride) as follows from calculations for 25 accident cases by Umeoka et al. [99Ume]. Maschek et al. showed that for a pure PuN core of CAPRA design re-criticality could occur in that case due to relocation of molten Pu [96Mas].

Nevertheless, nitride fuel remains an attractive option for innovative reactors if lead or gas is used as coolant (no FCI risk) and if an intrinsic safety approach (no CDA) is adopted. In that case the main concern is the limited thermal stability of AmN. Am redistribution in the fuel can be expected during irradiation, especially in case of high linear power. Increased gas pressure may lead to pin failure, and, if in the extreme case, nitrogen gas leaves the active part of the pin, positive reactivity feedback. Using ^{15}N enriched nitrogen, the nitrogen void worth is about 1000-1500 pcm smaller than for natural nitrogen. Combined with a large pin pitch, ^{15}N void worths can become negative, but can be accommodated within the typical sub-criticality margin of an ADS [2000Wal2].

As the relatively low(er) thermal stability of the actinide nitrides forms one of the major disadvantage of uranium-free nitride fuel, the addition of an inert matrix to improve the thermal properties of MA-based nitride fuel has been suggested. ZrN is seen as potential support. It forms a continuous series of solid solutions with UN above 800°C. The thermal conductivity of, e.g. $U_{0.5}Zr_{0.5}N$ is estimated to be $\lambda \approx 22$ W/(K · m) at 1000°C and $\lambda \approx 24$ W/(K · m) at 2000°C. Also AlN has been suggested as diluent.

Two designs for nitride fuels have been investigated: helium-bonded and sodium-bonded fuels pins. The interest of a sodium bond is obvious: it reduces the temperature difference between the fuel and the clad. Thus it is possible to design a pin with an initial fuel-clad gap large enough to accommodate the fuel swelling up to the target burnup and to operate the fuel at high linear power while keeping its maximum temperature low. In spite of this, the fuel designs have converged to a helium-bonded pin. Sodium bond led to complicated problems at the reprocessing stage because of its violent reaction with aqueous solutions. Of course, in case of pyrochemical reprocessing, this problem is no more a critical issue. Furthermore, it was feared that faults in the sodium bond (for example gas bubbles) could lead to hot spots and local melting. These faults could come from the fabrication and/or the irradiation (gaseous fission bubbles). However today extensive experience on the sodium-bonded metallic fuel exists in the USA (see Section 2.2).

With a helium bond, the poor thermal exchange between fuel and clad leads to a higher fuel temperature (above the "critical" point) at beginning of life which leads to higher swelling rate. The fuel-clad gap is rapidly closed and mechanical fuel-clad interaction (MFCI) will occur, and could limit the burnup to be reached. However, it is believed that such a mechanical interaction could be withstood by the clad up to the target burnup, provided that the pin smeared density and the cladding materials are optimised. Low fuel bulk density is required (80-85% of the theoretical density). Unfortunately the NIMPHE experiment, which was designed to demonstrate this (up to 15 at% burn-up), was never completed.

For (Pu,Am)N additional swelling can be expected due to helium production from in-pile decay of ^{242}Cm. Helium diffuses at lower temperatures than fission gases, and it may thus be impossible to avoid high swelling rates by lowering of temperatures. The remedy proposed is release of all helium produced by means of high extent of prefabricated porosity, which is normally the case for nitride fuels [99Pil]. Since this solution requires very large plenum volumes, a complementary approach may be introduction of neutron absorbers into fuel assemblies that would remove slower neutrons and hence suppress effective Am neutron capture cross sections [2000Wal1].

2.4.2 Fabrication

The existing know-how on the fabrication of nitride fuel is valid mainly for mixed (U,Pu)N fuel. While in the past (U,Pu)N fuels were fabricated by directly nitriding the metal with $N_2(g)$ or $NH_3(g)$ or by nitriding the actinide hydride, carbothermic reduction has become the standard method. In the latter process oxide powders are mixed with carbon, compacted and heated in a stream of nitrogen to produce a single-phase nitride. This product will normally contain some oxygen and carbon impurities. The classical procedure is then to mill the compact into a fine powder that is then pressed into pellets and sintered at about 1900 K. Alternatively, "direct pressing" of the compacts from the carbo-thermic reduction process allows to suppress the milling stage [91Bla]. The high reaction/sintering temperature of the nitrides forms a specific problem for MA-containing nitrides as AmN containing material has, as mentioned before, a limited thermal stability.

Another procedure utilises the sol-gel method, which was implemented at PSI for plutonium fuels. Here, actinide nitrates are mixed with an aqueous solution containing a gelating agent, urea and dispersed carbon-black. The mixture is dropped into hot silicon oil, which triggers the precipitation of actinide oxide microspheres that after washing can be subjected to carbothermic reduction in flowing nitrogen gas [88Ald, 92Led]. The composition for dense microspheres tends towards a carbonitride, with a low oxygen content. The microspheres may be pressed into pellets, or transferred directly into fuel pins by vibro-filling forming a fuel pin with the required low density to accommodate the high fission gas produced and fuel swelling. Densities between 66 and 84%TD are achievable depending of the sphere size fractions used.

The fabrication of the transuranium mononitrides has been studied extensively at JAERI (Japan). NpN, (Np,Pu)N, PuN, as well as small amounts of AmN and (Pu,Cm)N have been manufactured [99Ara, 99Tak,2001Tak]. Starting with the intimate mixtures of oxides, nearly complete solid solutions of various mixed-nitrides were obtained by the carbothermic synthesis.

As mentioned before, zirconium nitride is a candidate as diluent for uranium free nitride fuel fabrication. ZrN forms a solid solution with the actinide nitrides and is soluble in nitric acid [99Kle,2001Bur]. A problem that must be overcome is the high oxygen affinity of zirconium. Oxygen content in the fabricated nitride should not exceed one molar percent, if formation of an

oxide phase is to be avoided. For that reason (Pu,Zr)N fuel pins have been fabricated in by reaction of the metal with nitrogen gas at VNIINM (Moscow) [98Zab]. Difficulties are also expected when mixtures of ZrO_2 and AmO_2 are used to obtain (Zr,Am)N, since ZrC is very stable and removing carbon from ZrC would not be so easy without a significant loss of Am. One may have a choice of using the carbonitride (Zr,Am)(N,C), however. At JAERI a series of solid solutions of (Y,Am)N has been prepared by the carbothermic synthesis, since carbide of yttrium is less stable.

2.4.3 Irradiation behaviour

Experience from (U,Pu)N fuel

The Russian fast-neutron test reactor BR-10 has been operating on UN fuel for two full core burn-ups, with maximum burn-up reaching 9 percent. No data are available for uranium free nitride fuels, but a (Pu,Zr)N pin is expected to be irradiated in BOR-60 by the end of 2000 in the frame of the CAPRA programme.

Several irradiation experiments of UN and (U,Pu)N have been performed in Europe: the NILOC experiments [91Bla], the GOCAR experiments [84Ron] the NIMPHE experiments [92Fau]. In addition irradiation data are available from experiments in Japan [97Ara] and Russia. However the available data are not conclusive and uncertainties still exist for (U, Pu)N behaviour under irradiation. In particular, the effect of the fabrication process on the porosity distributions should affect significantly the swelling and gas release rate. Complementary knowledge comes from the experiments on carbides (see section 2.5.3). It is noticed is that nitrides swell less than carbides. Up to moderate burnup, the fuel "free" swelling (without MFCI) depends strongly on the fuel temperature. At low temperature (<1450-1550K), it does not depend on temperature and is closed to 1.6at%. That so-called "athermal" swelling is due to precipitation of the gaseous fission products, either as solution or as microbubbles. At this stage, fission gas release is rather small (about 2.5%) and independent of the burnup. At higher temperature, the swelling increases rapidly and depends exponentially on the temperature. This is the "thermal" swelling. This abrupt behaviour change, above a "critical" temperature, is mainly due to the observed migration to the grain boundary, coalescence and growth of the bubbles. The critical temperature decreases slowly with the burnup and depends also of the impurity concentration (oxygen and carbon for nitrides). At this stage, the gas release increases rapidly with the burnup. At high burnups, one observes a saturation of the free swelling because of the interconnection of the pores and a large release of the gaseous fission products, which could reach up to 70-80%. Furthermore, as soon as the fuel-clad gap is closed, the fuel cannot swell freely and the "restrained swelling" is lower than the free swelling. At the end of irradiation the swelling is mainly due to the "solid" swelling (coming from the solid fission products), which is constant (about 0.4% per at% burnup, similar to the oxide [71Bau]).

TRU fuel

No irradiation data exists for transuranium nitride fuels.

2.4.4 Summary

Nitride fuel has good thermal conductivity and the actinide nitrides show a mutual miscibility in a wide range of compositions. Nitride fuel is expected to have a good behaviour during normal operating conditions, especially when U-based. Uranium-free nitride, however, should operate at low temperature to decrease as much as possible the swelling of the fuel and also the Am redistribution resulting from vaporisation. Provided that design options are well defined (preferably sodium bond, low bulk and smeared density, ...) it could be possible to reach high burnup.

Irrespectively of the pin design, it seems difficult to avoid the consequences of the nitride vaporisation at high temperature in case of severe accident (CDA) because the expected complete dissociation of the metal nitride will lead to formation of N_2 gas (which leads to the over-pressurisation of the reactor vessel and to positive reactivity feedback) and actinide metal (which melt or volatilise at low temperature). The choice of nitride fuel for ADS thus requires very specific fuel and core design in order to be able to safely operate the fuel.

Nitride fuel requires enrichment of nitrogen in ^{15}N to avoid the ^{14}C production, which makes the fabrication process more demanding and has an impact on the fuel costs.

2.5 Carbide fuel

2.5.1 Characteristics

The transuranium carbides have the following characteristics:

- NpC_{1-x} (NaCl-type structure) is hypostoichiometric and exists between C/Np = 0.82 and C/Np = 0.96. The melting point is > 1673 K (1400°C). Complete miscibility with UC is probable. Further, the carbides Np_2C_3 and NpC_2 were observed.
- PuC_{1-x} (NaCl-type structure) is hypostoichiometric between C/Pu = 0.80 and C/Pu = 0.92 at room temperature and melts peritectically at 1654°C. The C richer phase Pu_2C_3 melts peritectically at 2050°C. PuC_2 is a high-temperature phase. The partial pressure of Pu above PuC_{1-x} - Pu_2C_3 is quite low: $p(Pu) = 2 \cdot 10^{-10}$ bar at 1000°C, $p(Pu) = 3 \cdot 10^{-6}$ bar at 1500°C. The thermal conductivity of PuC_{1-x} is fairly high: λ = 11 W/K·m at 1000°C, λ = 12 W/K·m at 1200°C. UC and PuC form a continuous series of solid solutions up to 70 at. % PuC_{1-x}. However, the phase becomes hypostoichiometric above this concentration limit. The Pu partial pressure over $U_{0.85}Pu_{0.15}C$ is $p(Pu) = 3 \cdot 10^{-8}$ bar at 1500°C. The thermal conductivity of $U_{0.85}Pu_{0.15}C$ is λ = 21 W/K·m at 1200°C.
- Am_2C_3 (bcc structure) has a high melting point. It has been prepared by arc melting. The existence of AmC_{1+x} (NaCl-type structure) is questionable. There seems to be limited solubility

of AmC in (U, Pu)C. As the free energy of formation of $AmC_{1.5}$ is only about -70 kJ mol^{-1} [84Hol] a comparatively high vapor pressure of Am at high temperatures during fabrication.

The technical information on carbide fuels is limited to uranium and plutonium carbide and can be summarised as follows. PuC and Fe are not in thermodynamic equilibrium. However, the reactions are kinetically hindered up to 800°C. In contrast, no reactions between UC and steel cladding materials were observed up to 800°C. However, UC reacts with Ni-base alloys above 600°C. With respect to compatibility with coolant it is known that No reaction of PuC with Na was observed. The same is true for UC up to 870°C. No reaction of UC with Pb or Bi was observed up to 950°C. The solubility of UC_2 in liquid Pb at 1100°C (2 hours dissolution time) was found to be 0.02 at. % U and 0.10 at. % C. Similar results can be expected for UC. Direct aqueous reprocessing of carbide fuels has been shown to be difficult. For example, UC, PuC and (U,Pu)C are dissolvable in concentrated HNO_3 and most of C is converted to CO_2. However, also organic acids are formed, e.g. mellitic acid $C_6(COOH)_6$, which interferes with the extraction step of wet reprocessing of irradiated UC. This can be avoided by dry conversion of the irradiated UC to UN before reprocessing.

A major drawback of the carbide fuels is their pyrophoric nature. PuC powder is pyrophoric at room temperature and reacts with nitrogen at 1400°C to form PuN and Pu_2C_3. Also UC powder is pyrophoric at room temperature.

A potential homogeneous diluent for carbide fuel is ZrC. It forms a continuous series of solid solutions with UC. The thermal conductivity is high, e.g. $U_{0.5}Zr_{0.5}C$: λ = 17 W/K · m at 1000°C, λ = 23 W/K·m at 2000°C. The mutual solubilities of PuC and ZrC in the PuC-ZrC system are, however, limited below 1600°C.

2.5.2 Fabrication

The existing know-how on the fabrication of carbide fuel is restricted to mixed (U,Pu) carbides. Considerably more experience exists compared to nitride fuel, and mixed carbides are the most well developed of the alternative ceramic fuels. Large scale experience with the production of mixed carbides has been gained at Los Alamos laboratory in the US [83Mat], where a glove box line was constructed with a capacity of 1000 pellets per day for test pin production, and in the Babha Atomic Research Centre in India, where $(Pu_{0.7}U_{0.3})C$ fuel is used as driver fuel in the Indian FBTR. The processes used in these installations are based on the carbo-thermic reduction of the oxides which is nowadays to be preferred to the earlier used methods of reacting uranium metal with carbon or methane. In the carbothermic production process pre-compacted mixtures of the oxides and carbon are heated in vacuum above 1300 °C. The compacts are then crushed, milled and pressed into pellets, which are sintered. The M_2C_3 content in the fuel generally must be limited to less than 10%, which can be achieved by heating in hydrogen. Because the powders of the actinide carbides thus produced are very pyrophoric, an inert glove box atmosphere (argon) is required for the fabrication and the oxygen content in the gas should be kept low (< 10 ppm). This does not only

apply to the pellet fabrication but also to the subsequent fabrication steps and scrap recovering and recycling. The handling of powders and pressing of pellets can be avoided when a carbothermic reduction process based on the internal gelation technique is used, as was developed by PSI. Carbon black was introduced into the gelation agent solution and the dried microspheres were reacted to dense carbide microspheres. Two size fraction filled into a cladding tube give smeared density similar to the requested density for high burnup application.

However, little is known about the production of uranium-free carbides. Plutonium carbides (PuC, Pu_2C_3) and americium carbide (Am_2C_3) have only been produced in small quantities for research purposes.

2.5.3 Irradiation behaviour

Experience from (U,Pu) fuel

There is extensive information on the irradiation behaviour of mixed (U,Pu) carbides. More than 5000 fuel pins of (U,Pu)C have been tested in irradiation experiments, including ramp tests. Two different pellet fuel concepts have been investigated: liquid metal (essentially sodium) and helium bonded. Because the early results from irradiation experiments indicated that, in spite of the good thermal conductivity, carbide pellet fuel can only be operated at low linear ratings (due to the fission gas induced swelling), there is not a big distinction between the two concepts from the point of view of fuel behaviour and experiments have provided no arguments to favour one of them. Sodium bonding was regarded more reliable but also more expensive. In addition to these concepts, helium bonded particle fuel has been investigated.

In the US an extensive irradiation programme was performed in the EBR-II fast reactor. It was found that a low smeared density of the fuel is required to avoid fuel-cladding interaction as a result of the swelling of the fuel with increasing burn-up. The maximum allowable pellet density was found to be ~85 %TD. In EBR-II He-bonded test pins containing (U,Pu)C of low smeared density in AISI 316 stainless steel cladding were irradiated up to 20.7 at% burn-up without failure. A full size bundle of 66 pins with pelletized fuel and 25 sphere-pac pins was irradiated as test AC-3 in the Fast Flux Test Facility (FFTF) with a linear heating rate of 80 kW/m up to a burn-up of 8 at% in a D9 cladding [93Str]. The microspheres with 20% Plutonium and the sphere-pac fuel pins were fabricated at PSI [88Led]. Excellent behaviour was found with no cladding carbonisation and an indication of a homogeneous load on the cladding with sphere-pac fuel.

Also in Europe (U,Pu)C fuel with low smeared density have been tested extensively, mainly in programmes in France, Germany and UK:

- In France fuels with a smeared density of 71% was successfully tested in irradiation experiments in thermal reactors up to 120.000 MWd/tHM.

- In Germany fuels with a smeared density of 75% which were tested successfully in the KNK-II fast reactor up to 9 at% and in power cycling and overpower tests. Also a comparative irradiation test of pellet and sphere-pac fuels was made in BR-2.
- In UK fuel with a smeared density of 70% was tested, with the aim to reach a burn-up of 100.000 MWd/tHM. The UK researchers, however, favoured the sphere pac (helium bonded) concept, which was originally developed by EIR (now PSI) in Switzerland. In this concept the sol-gel technique is used to produce spherical particles of two sizes which are loaded in a fuel pin by vibrocompaction.
- A Belgian-Swiss irradiation test (MSBF7) of sphere-pac and pellet carbide fuels has been performed in the BR-2 reactor, which indicated less cladding strains and less pin failure for the sphere-pac fuels [80Del].

At present India is the only country with an on-going carbide irradiation programme. In the Indian FBTR $(Pu_{0.7}U_{0.3})C$ has been used as driver fuel since 1985 with success. The fuel has reached a burn-up of 50000 MWd/tHM without any failure. The fuel will be replaced progressively with $(Pu_{0.55}U_{0.45})C$ fuel in the coming years.

TRU fuel

No irradiation data are available for uranium-free carbide fuels.

2.5.4 Summary

Uranium and plutonium carbide fuels have good thermal and mechanical properties and have shown good irradiation performance. Helium bonding was found to be sufficient to reach high burnup. Unlike the nitride fuel, there is no safety concern regarding the case of severe accident (CDA). The highly pyrophoric nature of the carbide compounds requires an inert atmosphere for fabrication. In case of transuranium carbide fuel, a strict control of stoichiometry is required in order to obtain a monocarbide phase. The compatibility with wet reprocessing is only given by transforming the spent fuel to a carbonitride. Overall, this fuel type is considered less interesting than most other fuels described in this report.

2.6 Composite fuels

2.6.1 Ceramic-ceramic fuel (CERCER)

2.6.1.1 Characteristics

CERCER composite fuels/targets for actinide transmutation have been studied extensively in the recent years [98Coc]. Especially in the frame of the EFTTRA collaboration and the French SPIN programme different fuel concepts have been tested. These concepts can be divided in micro- and macro-dispersions. In a micro-dispersion the actinide phase is distributed as small particles of undefined shape (< 30 μm) in the matrix, in a macro-dispersion spherical particles of a size between 50 and 300 μm are distributed in the matrix. Clear differences are to be expected in the irradiation behaviour of these two concepts, especially with respect to the effect of damage of fission products. In the micro-dispersion the damage caused by stopping of the fission fragments in the matrix is distributed almost homogeneously in the pellet. In case the matrix is very susceptible to this damage this will lead to deterioration of the properties of the fuel during irradiation. In case of a macro-dispersion, the damage is restricted to a small shell around the particle. Thus less than 10% of the matrix will suffer from the effects.

Of course, the ceramic matrix must fulfil many other criteria such as sufficiently high melting point, good radiation stability, good mechanical properties, compatibility with coolant, etc., and only few materials have been found to be attractive. Magnesium oxide (MgO) is the main candidate for fast reactor applications. Also magnesium aluminate spinel $MgAl_2O_4$ has been suggested and subjected to in-pile and out-of-pile tests, but the radiation stability of this material is much less. For the dispersed phase the mixed actinide oxide is a logical candidate but also the YSZ-based phase is considered, especially in case of Am-rich fuels.

As noted in section 2.3, homogenous oxide fuels have poor thermal properties, limiting their application. CERCER fuels consisting of an actinide host phase and a ceramic matrix with good thermal conductivity allow to improve the thermal properties of the fuel significantly. When $(Pu,MA)O_2$ is dispersed in MgO in a 1:2 volume ratio, it is possible to improve the thermal conductivity of the fuel by a factor of two at beginning of life. This may be enough to keep a sufficiently large margin to the fuel melting with conventional pin geometry (without central cooling channel) and probably to design a fuel pin compatible with gas coolant.

It is important to note that in spite of the extensive R&D efforts on composite fuels/targets, many data are still missing to model with confidence the behaviour of such fuels under normal or accidental reactor operating conditions. For example, the possibility of eutectic melting at temperature significantly below the melting point of the two phases has been reported [2001Sch] for $MgAl_2O_4$-$(Zr,Y,Er,U)O_2$ composite fuels. Also information about swelling and gas release versus

burn-up, thermal conductivity degradation with burn-up, compatibility with Pb or Pb/Bi, etc. is not known.

2.6.1.2 Fabrication

Most experience gained for CERCER fuels or targets so far, is based on UO_2-based materials. Some experience exists with Pu-based or Am-based materials. The main fabrication process of composite fuel pellets is based on powder metallurgy, where the powder of the dispersed phase can consist of a fine-grained powder or of spherical particles of the actinide phase. The powders are generally mixed together or obtained by co-precipitation from a nitrate solution. An alternative process is the infiltration of porous pellets or sol-gel beads of inert matrices by a nitrate solution of actinide. This method allows simplification of the process steps to be performed in glove box environment and avoids all the problems arising from handling of powders. Combinations of infiltration and powder metallurgy are also possible, in which spherical particles of a host material (e.g. stabilised zirconium oxide) are infiltrated with the actinide, and next mixed with the inert matrix. However, the development costs of this type of fuel may be considerable, if not prohibitive, compared to the more simple homogeneous fuel.

At CEA (Atalante Laboratories) the fabrication of MgO-AmO_{2-x} microdispersed fuel has been undertaken successfully [2000Cro]. Powders of MgO and AmO_{2-x} (with $x \approx 0.4$) have been blended, pressed and sintered, to achieve pellets of high density (> 95%). Also macrodispersed fuel containing plutonium have been fabricated on a laboratory scale. In a collaboration of JAERI, NRG and PSI macrodispered fuel samples of $(Zr,Y,U,Pu)O_{2-x}$ spheres, produced by internal gelation, in $MgAl_2O_4$ have been fabricated [2001Sch]. The infiltration of spheres with actinide solutions, followed by blending with $MgAl_2O_4$ or MgO powder is being studied at ITU [2000Bou]. The problems that have been met in the these fabrication studies are the difficulties to achieve a homogeneous distribution of the dispersed phase, to assure a good thermal interface between dispersed phase and avoid the protrusion of the dispersed phase.

Fabrication of dedicated fuels at a larger scale than that of a laboratory should meet the same difficulties as described earlier.

2.6.1.3 Irradiation behaviour

MgO-PuO_2 fuels were investigated in the ETR the USA in the 1960s [63Fre]. The results showed, in general, satisfactory performance, in spite of the extreme conditions of linear rating (1650 W/cm) and extent of fission (72% FIMA). Fuel samples with 2.71 and 12.95 wt% PuO_2 in MgO (macro-dispersion) showed that pellet cracking and gas release were smaller than in samples containing UO_2, tested under the same conditions. In the sample with the highest linear power a central hole was formed and columnar grain growth occurred as is known for $(U,Pu)O_2$ fuel in fast reactors. At the same time, a significant redistribution of plutonium (a factor 100 in concentration between

inner and outer ring of the pellet) was found. In spite of the high linear ratings, the central temperature was not extremely high, because of the good thermal properties of magnesia.

In the EFTTRA-T4 experiment a ceramic-ceramic (CERCER) target type composed of americium oxide dispersed in magnesium aluminate spinel ($MgAl_2O_4$) was irradiated in the HFR at Petten during 358.4 full power days [2000Kon]. During the irradiation the ^{241}Am concentration was reduced to 4% of its initial value. The fraction of the initial americium atoms that had been fissioned was 28%. Post-irradiation examinations showed that the target swelled considerably during the irradiation (up to 18%) which is caused by accumulation of helium, the product of the alpha decay of ^{242}Cm that is produced in the transmutation scheme of ^{241}Am.

Other experiments on dispersion fuels in the EFTTRA and SPIN programmes were made with enriched UO_2 instead of minor actinides. In these samples the effect of helium is absent but the radiation damage should be comparable. The results of these experiments (TANOX, EFTTRA-T3, MATINA-1, THERMHET) are, however, not conclusive with respect to the advantages and disadvantages of the macro- and microdispersion concept and good and bad results were obtained for both concepts. Additional work is needed to understand these contradictory results.

In the research programme of JAERI so-called rock-like fuel (ROX) was studied, mainly for plutonium incineration. In the first concept the polyphase mixtures $(Zr,Y,Pu)O_2$+$MgAl_2O_4$+Al_2O_3 and $(Th,Pu)O_2$+$MgAl_2O_4$+Al_2O_3 were prepared [99Yam]. Samples were irradiated in the JRR-3 reactor to a burn-up of 21-28%. These samples did not behave very well: significant swellling and reactions between the phases were observed, which was attributed to the presence of Al_2O_3. In the later concept, the ROX design was changed to a two-phase mixture of $(Zr,Y,Pu,U)O_2$ and $MgAl_2O_4$. An irradiation of this material in collaboration with NRG and PSI is in preparation [2001Sch] whereas the uranium analogue was tested in JRR-3 [2001Kur].

2.6.2 Ceramic-metal fuels (CERMET)

2.6.2.1 Characteristics

In ceramic-metal (CERMET) fuels a ceramic actinide phase, generally an oxide, is dispersed in a metal matrix in order to combine the main advantages of ceramic and metallic fuels:

- The metal matrix guarantees a high thermal conductivity of the fuel as a result of which the operating temperature is low. For example, the thermal conductivity of a CERMET in which volume ratio of the metal is 0.66, is a factor of 30 better than that of UO_2 fuel at beginning of life, compared to a factor of 2 in a CERCER fuel of the same matrix/actinide ratio.
- The actinide-containing oxide phase is more easily to fabricate and to handle compared to the actinide metals as it contains the minor actinides in a stable oxidation state.
- A dense metal matrix can help to retain fission gases, volatile fission products and helium in the fuel.

The most interesting metal matrices for CERMET fuel are steel, V, Mo, Cr, Zr or Zircaloy considering the neutronic aspects. However, the neutron-capture cross sections of some of these metals are not insignificant and the activation needs to be analysed in more detail. For example, activation of Mo will lead to the formation of technetium, which is one the fission products that is considered for transmutation.

2.6.2.2 Fabrication

The fabrication of CERMET fuel is generally done by powder metallurgy. The metal powder is mixed with the ceramic phase, pressed into pellets, preferably by cold compaction, and sintered at high temperature. The problem to be solved is the control of the right distribution of brittle ceramic phase in the ductile metal matrix. As is the case for CERCER materials, a spherical size of the ceramic phase is considered ideal and stabilisation of the actinide in a host phase (e.g. stabilised zirconia) is also preferred.

The existing information on the fabrication of CERMET fuels is limited to uranium oxide as actinide phase of inactive phases to simulate the actinides. In the 1960s work on the fabrication of CERMET has been performed in the US where UO_2 was dispersed in metals iron, niobium, aluminium, zirconium or alloys like stainless steel (austenitic and ferritic) or Zircaloy. When working with ductile metals that are not very sensitive to oxygen (e.g. Mo), the fabrication is relatively simple and can be based on powder mixing. However, for zirconium, an interesting metal for CERMET fuel, this is not the case. For that reason, more complex fabrication techniques are investigated. In Russia Zr-UO_2 CERMET fuel has been fabricated from zirconium powder and UO_2 powder using a hot extrusion process [2001Tro]. Alternatively, the fabrication of Zr-based CERMET fuel can be achieved using zirconium hydride (ZrH_2) as starting material, which is mixed with the ceramic phase and converted to metal by thermal treatment. Such fabrication processes are still in the initial phases of research.

An innovative fabrication method for oxide CERMET fuel was developed at Kernforschungzentrum Karlsruhe (now FZK) in the 1970s [73Wei]. In this process an oxide kernel (UO_2) was coated with a metal (Cr or V), the coated particles were vibrated into a cladding tube (Inconel or Hastelloy), after which the welded tubes were hot-isostatically pressed under helium. CERMET fuel with 20-30 vol% metal were fabricated, having an idealized fuel structure (continuous metal network, no gap, no plenum, density about 85% of the theoretical one) and a metallurgically bonded cladding.

2.6.2.3 Irradiation behaviour

Experience from (U,Pu) fuel

As already mentioned, pioneering work on CERMET fuels was performed in the 1960s, with UO_2 as ceramic phase. Dispersion fuels of (stainless steel - 25% UO_2) and (aluminium - 54% UO_2) were irradiated successfully to high burn-ups in material testing reactors, though it was observed that the localised increases in fission density and fission product concentration may cause stresses and irradiation instability [57Whi]. Oxide CERMET fuels rods with idealised structure of KFK design, as described in the previous section, were irradiated in FR2 up to 95 MWd/kgU [73Wei]. CERMETs of UO_2-30%Cr in Inconel-625 were irradiated successfully. At the same burnup, Hastelloy claddings were damaged by microcracks. Rods with UO_2-20%Cr in Inconel-625 indicated vapour transport of Cr from centre of the rod to the outer regions as a result of the higher central fuel temperature.

More recently, irradiation experiments of Mo+UO_2 fuels were performed by CEA (France). (TANOX-1 in Siloé and MATINA-1 experiments on Phénix). The results indicated excellent fuel behaviour with a low fuel temperature and low fission gas release [95Deh].

TRU fuel

CERMET fuels with a ceramic phase containing only transuranium elements have not been studied. Compared to UO_2-based CERMETS, the gas production in minor-actinide containing fuels, the helium production will be much higher and must be taken into account as this is also localised.

2.6.3 Metal-metal fuels (METMET)

The metal-metal type fuel is a variation of composite fuel for which relative little is known. It has been proposed recently in the US ATW proposal as the primary candidate fuel form [99DOE]. A dispersion of 20-40 μm particles of 25%Zr and 75% TRU (mostly Pu) in a matrix of Zr is proposed (overall fraction of TRU is 25%). This fuel is expected to have a good high-burn-up irradiation performance and excellent shock resistance in the event of accelerator beam interruption. As a mixed transuranium alloy obtained from the electrorefining can be used, a fabrication route may not require high-temperature processing. Possible fabrication methods, as outlined in the ATW report, involve adjusting composition of the alloy (increasing the Pu content), crushing the alloy (eventually in combination with hydriding), blending, pelletization and sintering. The feasibility of all these steps has to be investigated.

2.6.4 Summary

Composite fuels (ceramic-ceramic, ceramic-metal and metal-metal) have the advantage that the properties of the fuel can be tailored to a certain extent by a careful selection of materials. For ceramic-ceramic fuel types MgO is the most likely inert matrix host, AnO_{2-x} or $(Zr,An)O_{2-x}$ the most likely host phases. The number of metal matrices is also limited, and insufficient information is available about their compatibility with host phases and their behaviour under irradiation, expect perhaps steel, molybdenum and zirconium metal. They could be combined with $(Zr,An)O_{2-x}$ in a ceramic-metal type and with (An,Zr) alloy in a metal-metal type. It should be noted that the $(Zr,An)O_{2-x}$ oxide phase is considered to be a better than the (An,Zr) metal phase. The fabrication of composite fuel is considerably more difficult than for homogeneous fuels, especially in case of specific requirements for the size and distribution of the dispersed actinide phase. The development costs of this type of fuel thus may be considerable, if not prohibitive, compared to the more simple homogeneous fuel. Limited experience exists in this field. Also the irradiation behaviour forms a major uncertainty, especially the effect of locally high production of helium and fission gases on the mechanical behaviour.

2.7 Coated particle fuel

2.7.1 Characteristics

Coated particle fuels have been designed in the past for "high temperature reactors" (HTR). The interesting aspects of such fuel are: (i) the good thermal exchange between the gas coolant and the fuel as a result of the small fuel particles, and (ii) the retention of the fission products within the particle as a result of the tight coatings surrounding the fissile kernel. Experience has been gained with different types of kernels (UO_2, PuO_2 and ThO_2 as well as UC_2) in about 10 countries which performed during 20 years (1960-1980) all the research and development studies needed to prove the feasibility of such fuels for HTR and to qualify the behaviour under irradiation in thermal flux conditions.

The particle is composed of a spherical fissile or fertile kernel (typically oxide, carbide or oxycarbide of enriched uranium, fertile uranium and plutonium or thorium) of about a few hundred μm. The kernel is coated by one layer of porous pyrocarbon (PyC) of 80-100 μm of thickness, and several other dense coatings. For a BISO-type fuel (in principle designed for the fertile fuel element $^{238}UO_2$ or $^{232}ThO_2$), the kernel coatings consist in only one layer of dense PyC. For a TRISO-type fuel (fissile elements), two layers of dense PyC between which one layer of silicon carbide (SiC) is needed to ensure good mechanical properties and to ensure the fission gas retention. (For higher temperature applications ZrC is considered.) The thickness of each dense

layer is about 30-50μm. Coated particles are then embedded either in compacts (small full or annular cylinders) or in pebbles, both in graphite. For fast flux conditions these coatings materials have to be re-assessed in order to assure their resistance to the different irradiation conditions. It is not very likely that the PyC layer can withstand high fast neutron fluence. As a combined result of irradiation shrinkage and creep, fine cracks can be formed in PyC, which causes gas permeability. Once the inner-PyC becomes gas permeable, the SiC layer will suffer from the corrosion by CO.

A problem that deserves to be analysed in-depth is the dilution ratio (fissile volume/diluent materials volume) which is necessarily low when the fissile kernels are coated and embedded in diluent materials. As example, the ratio is about 1/1000 for a HTR core and 1/10 for a gas-cooled fast reactor. The efficiency of a diluted core to transmute minor actinides will have to be assessed compared to core using more conventional fuels.

2.7.2 Fabrication

The fabrication processes optimised for HTR particle fuels are divided in 3 steps:

1. Kernel fabrication. Different processes have been tested for the kernel fabrication: UC_2 fusion, calcination of carboxylic resins loaded with uranium, SOL-GEL or slow growing. The SOL-GEL process became the industrial reference. It allows fabrication of well-graded particles with high density and resistant to attrition. This latter property is important as it ensures the good quality of the coating deposit since it is necessary to avoid kernel pollution with dust coming from the mechanical shocks of particles.
2. Coating. The coatings are deposited by Chemical Vapour Deposition (CVD with acetylene, propylene, methyltrichorinesilane and methane gases). Special furnace, called coaters, were developed: size and form of the furnace, number and distribution of the nozzles, gas composition and flow, temperature, etc. were optimised to meet all the fuel specifications.
3. Fabrication of compacts. The compacts are made by mixing the coated particles with graphite. The mixture is pressed by hot-isostatic pressing into spherical compacts (German concept) or casted into rodlets (US/Japanese concept).

The fabrication of particle fuels has been undertaken on a large scale in Europe in the past for the German HTR programme. Since this programme has been stopped, the facilities have disappeared slowly. At the present time no facility is available in Europe. However, within the 5th Framework programme of the EU, the (still) existing knowledge is being revived and also some experimental research in kernel fabrication and coating technology will be started.

2.7.3 Behaviour under irradiation and modelling

Experience from (U,Pu) fuel

The good irradiation behaviour of particle fuel was demonstrated in tests that were successfully performed in experimental reactors for the following conditions: helium temperature 800-900°C, fuel temperature 1200-1400°C, burn-up 10-20 at%, and fluence up to 5 10^{21} n/cm^2 (E>0.18 MeV). The majority of these tests were performed in thermal reactors.

Few experimental data are available concerning the behaviour of particle fuels in a fast flux: only two irradiation experiments in the framework of Gas Cooled Fast Reactor were performed: one in DFR, the other in Rapsodie in the 1970's. Coated particles (embedded or not) of enriched uranium oxide or mixed oxide were irradiated up to a relatively low burn-up of 2-3 at %. It has been shown that some difficulties could occur in fast fluxes (fissuring and loss of tightness of the coating, compacts swelling) and therefore an experimental programme has to be started to adapt the "HTR particle fuel technology" to fast reactors. Particular areas for attention are the coating composition, the reprocessing feasibility, the particle behaviour under irradiation, and the particle and fuel designs. The diffusion of certain fission products through the different layers was considered a limiting phenomenon for HTR fuels, which has to be analysed again in the fast reactor conditions, taking into account the fuel temperature, the target burn-up, etc...

TRU fuel

No experience exists with pure transuranium coated-particle fuels. Compared to the HTR fuel, the behaviour under irradiation of MA-based particle fuels will be worsened by the large gas production (helium + fission gas). The design of the particle (number of layers, thickness of the porous and dense coatings) will have to take into account the thermal-mechanical effect of such a production on the resistance of the coatings to rupture. Furthermore, the kernel swelling risk is increased and could contribute significantly to the mechanical stresses on the coatings. Thus, the proposal is made to dilute the fissile kernel with compounds that are able to form a solid solution with the fissile compounds, as for oxide or nitride pellet fuels, with the objective of improving the thermomechanical stability of the kernel.

The possible chemical interaction of the MA oxide kernel (if that is the case) with the carbide-type coatings will have also to be examined as it can lead to the additional formation of gaseous carbon monoxide CO. Since the fission yield of Pd is one order of magnitude higher for TRU than U-235, the Pd attack on SiC has to be studied closely.

2.7.4 Summary

Coated particle fuel is essentially a special form of composite fuel. It is well known from the HTR development and offers the possibility of gas retention and would thus be suited for MA fuels with high gas production. However, new concepts need to be developed for a fast neutron system, as carbon will act as a neutron moderator. The choice of the coatings for such a system is a key issue.

3. FISSION PRODUCT TARGETS

The long-lived fission products that are most frequently considered in transmutation studies are ^{99}Tc, ^{129}I and ^{135}Cs. They can be transformed into stable isotopes by a single neutron capture.

Technetium metal is an excellent target material for transmutation in reactors. As part of the EFTTRA collaboration, irradiation tests of this material were performed in the High Flux Reactor (HFR) at Petten and an irradiation test in the Phénix reactor is in preparation [97Kon1, 98Kon, 99Kon]. The fabrication of the cylindrical targets for these tests was made by casting liquid technetium metal in a water-cooled copper moulds. The targets each contained two such cylinders. The EFTTRA-T1 and T2 irradiation tests showed good behaviour of the technetium in which about 18% was transmuted to the stable ^{100}Ru isotope. In the rim of the cylinders the extent of transmutation was considerably higher (30-40%) due to contribution of neutrons from the resonance region, where neutron capture cross section of ^{99}Tc is much larger than for thermal or fast energies, typical for LWRs and FNRs, respectively. To make use of this effect, experiments on the transmutation of ^{99}Tc were performed at CERN applying the adiabatic resonance crossing (ARC) method in lead where neutrons lose their energy in small decrements. This allows access to the resonance energy region and, thus, the efficiency of the transmutation process may be increased. The CERN researchers have shown that ^{99}Tc can be introduced in the lead volume of an ADS core in a parasitic mode, and transmuted without affecting the operation in a significant way [99Arn].

For iodine targets, metal iodides are being considered. An irradiation experiment to test the behaviour of some metal iodides was performed by the EFTTRA group [97Kon1,97Kon2]. Pre-test analysis showed that the candidate materials can be classified in groups of similar properties. CeI_3 and YI_3 were considered to be very promising materials, having a relatively low cross-section for thermal neutron capture, reasonably high melting point, a low vapour pressure and a high thermodynamic stability. However, they have the disadvantage of not being stable towards moisture and oxygen (they easily form oxy-iodides) and are hitherto difficult to prepare in a pure form. This can only be avoided by careful handling in an inert atmosphere. PbI_2 and ZnI_2 are much less stable in terms of thermal dissociation, have relatively high vapour pressure and the low melting points among the compounds considered, but are relatively easy to handle. MgI_2, CaI_2 and NaI have intermediate characteristics, and are therefore in most aspects less promising than CeI_3 or YI_3. In addition, the compatibility with PUREX reprocessing technology has to be considered in the choice of a suitable material: in case of iodine removal, NaI is the product of liquid scrubbing of off-gases, and PbI_2 is the product of the conversion of AgI from solid adsorbents. From the three groups CeI_3, PbI_2 and NaI were selected for the EFTTRA-T1 experiment, which was performed with the stable ^{127}I. In this experiment about 5% of the ^{127}I was transmuted to xenon. Post-irradiation experiments showed that CeI_3 and NaI had behaved well during irradiation but that PbI_2, which was in a molten state during the irradiation, had corroded the stainless steel cladding of the capsule. The overall conclusion of the EFTTRA study was that if transmutation of iodine is considered, sodium iodide is the best target material, although mediocre in many aspects.

Recently, an irradiation experiment on the transmutation of Iodine, code-named project I, has been performed at the HFR Petten [2001Sch2]. Taking into account the results of the EFTTRA-T1 iodine transmutation experiment, the following selection of metal iodides was made: NaI, MgI_2, CaI_2 and CuI. These metal iodides were irradiated in the form of pellets with densities over 89% T.D. The estimated extent of transmutation of these targets is in the range 6.6-7.3 at%. At the end of the year 2000 the irradiation was completed, and the post irradiation examinations have been started, but no data are yet available.

Caesium transmutation in reactors does not seem feasible from the point of view of the neutron economy. The neutron absorption cross section of ^{135}Cs is low and separated caesium is not a single isotope but a mixture of the long-lived ^{135}Cs, the short-lived ^{137}Cs and the stable ^{133}Cs, as a result of which parasitic neutron capture in especially ^{133}Cs will occur.

Summary

- **Technetium metal has been shown to be an excellent target material for transmutation of ^{99}Tc.**
- **Of the metal iodides studied, sodium iodide (NaI) is considered to be an acceptable target material of ^{129}I though its properties are mediocre. More details about the positioning and the thermal environment of the targets in the ADS (core, core periphery, lead) need to be known to make a more detailed analysis.**
- **Caesium transmutation does not seem feasible.**

4. REPROCESSING TECHNOLOGY

Two types of processes can be applied to the separation of long-lived radionuclides: hydrochemical ("wet") and pyrochemical ("dry") processes. Hydrochemical processes are very well suited for the processing of large quantities of low-enrichment fuels that have had long cooling times after discharge. They are thus used on a commercial basis for fuel processing of LWR UO_2 or MOX fuel for re-cycling of Pu, or to fabricate specific waste forms for disposal. Pyrochemical processes cannot be considered as an alternative to hydrochemical processes like the commercial PUREX. Because of the batch nature of pyrochemical processes, materials handling and transfer are important and at the present stage of development severely restrict the throughput rate for such processes. But if the quantities to be processed are small, or if the fissile content of the fuel to be processed is high, or if the discharged fuel has been cooled for less than one year, pyroprocesses are very useful. For these reasons dry processes may be very attractive to new reactor concepts for transmutation such as ADS. In this chapter of the report an overview of the R&D programmes on hydrochemical processes and the state of the art of the ongoing R&D programmes on pyrochemical processes is given.

4.1 Hydrochemical processing

4.1.1 Description of the PUREX and related processes

This paragraph provides a brief overview of the main hydrometallurgical processes, which could be used as partitioning technologies of actinides, mainly transuranium elements, for the following fabrication of fuels and targets with a view to their possible transmutation and recycling.

The PUREX process is the most important hydrochemical reprocessing technique to separate U and Pu from spent fuel containing natural, slightly or highly enriched uranium [49Lan,79Koc]. It is employed in the spent fuel reprocessing industry and is taken as reference process in the LWR/UOX and LWR/MOX reprocessing. It applies tri-n-butyl phosphate (TBP) in a kerosene-type aliphatic hydrocarbon diluent as the extractant and utilises the highly selective extractibility of tetravalent U and Pu nitrates to separate them from the bulk of fission products (FP) in a nitric acid (HNO_3) medium. The present state of the art in reprocessing allows the separation of U and Pu with effciencies of 99.88 % . The minor actinides (Np, Am, Cm) and the fission products remain in the liquid HLW and are incorporated in the vitrified high-level waste.

Three different options can be distinguished in the PUREX process according to which the partitioning of TRUs (Np, Pu, Am and Cm) can also be achieved:

Standard PUREX process:

U and Pu are separated with an industrial yield close to 99.9 %. MAs and FPs are conditioned in a glass matrix for interim storage and final disposal. Am and Cm are not separated because their valences (III) are not extracted by TBP. They remain in the raffinate (HLLW) of first cycle together with the bulk of fission products, except the gaseous like Kr-85 and I-129 which are separated in the off-gas treatment of the spent fuel dissolution. Np is partially extracted (~60 %) by TBP, following the U stream and it is separated from the latter in the second U purification cycle. This Np stream is added to the HLLW.

Improved PUREX process:

It is based on the standard PUREX process with the improved separation of Np. The chemical properties of Np are similar to those of U and Pu. Neptunium occurs in dissolution solutions feeding to first decontamination cycle as a mixture of Np (V) and Np (VI), but only the latter is extracted by TBP. The proportion of the two species may be modified by adjusting the Np valence in the dissolution solutions with an oxidising reagent to favour Np extraction by the TBP [93Uch,97Gui]. The Np is then extracted together with U and Pu in the first decontamination cycle and follows the U stream after Pu partitioning. Finally it is recovered through a reducing scrub in the second U cycle. After separation, the Np nitrate, contaminated by β-γ emitters, may be purified by solvent extraction with TBP and finally transformed to oxide by calcination of the oxalate. An other way is to follow the standard PUREX process to separate about 60 % of the Np from dissolution solutions. The remaining 40 % follow the raffinates (HLLW) of the first cycle. At this point, the inextractable Np (V) may be oxidised to Np (VI) or reduced to Np (IV), and is then extracted by TBP which is sent to the TPB stream of the first cycle prior to U/Pu partitioning. Therefore all Np will be found in the raffinates of the second U cycle. From a technical point of view this latter procedure is more complex to implement than the former, because it requires an additional extraction cycle of HLLW. However, from chemical point of view it has the advantage to carry out the Np valence adjustment without Pu presence. This allows the use of a wider variety of oxidising and reducing reagents. The recovery of Np achieving a 95 % yield has been demonstrated on laboratory scale and further improvements towards a 99 % recovery yield is under development. Other processes as DIDPA and TRUEX are researched to recover Np from HLLW. These and other processes are discussed with more detail below in the extended PUREX process.

Extended PUREX process:

This process is under research and development. It includes the separation of minor actinides (Am and Cm) and some fission products, mainly long lived. Special extractants are required for minor actinides (Am and Cm) separation from HLLW because their stable valence (III) is not extracted by TBP. Also these solvents will extract the lanthanides because their stable valence is also III. Three alternatives could be available to separate minor actinides (Am and Cm) from HLLW:

1. The first is based on co-extraction of minor actinides (III) and lanthanides (III). The developed or under development processes are:

- TALSPEAK (USA) and DIDPA (Japan), which use acidic organophosphorus as extractants [81Kub,84Per,94Wea,96Mor]. Also the DIDPA process could be used to recover Np from HLLW
- The TRUEX process (USA) is based on the use of CMPO (n-octyl-phenyl-di-isobutyl-carbomoylmethyl-phosphine-oxide) [88Sch].
- The TRPO process (China) uses a trialkyl phosphine oxide. This process has also been developed at the ITU (Karlsruhe) [94Zhu,94Son].
- The DIAMEX (diamide extraction) process has been developed at CEA (France) using malonamides as extractants [94Mad].

2. The second alternative is based on the separation of minor actinides in a single operation leaving all the lanthanides in the HLLW. High selectivity extractants like TPTZ (UK-France) (tripyridyltriazine) [85Mus] or CYANEX 301 (China) (trimethyl-pentyl dithiophosphinic acid) [96Zhu,99Mod]. The latter can be used in synergistic combination with TBP. New extractants and methods are under research and development. Within the 5th Framework Program of the EC two projects are under study: PARTNEW and CALIXPART. PARTNEW includes the development of solvent extraction processes for the separation of trivalent MAs (Am and Cm) from PUREX HLLW raffinates. The main extractants to be developed and tested are diamides (DIAMEX), nitrogen polydendate ligands, dithiophosphinic acids, and acidic S, Se or Te bearing ligands. The last three groups of compounds will be used on SANEX (Selective Actinide Extraction) process to separate selectively the trivalent actinides (Am and Cm) from HLLW. The objective of the CALIXPART is the synthesis of matrices able to extract and separate in a single step actinides from lanthanides. Several ligating chains including nitrogen or sulphur atoms properly organised on matrices such as calixarenes and others will be synthesised and tested.
3. The third alternative is the oxidation of minor actinides and their selective extraction. This is based on the fact that minor actinides, mainly Am, can be found in nitric solution at many degrees of oxidation unlike the lanthanides, except cerium, which are essentially trivalent elements. The direct separation of Am from HLLW presents two different points. The presence of ruthenium could perturb the oxidation process, therefore the ruthenium must be eliminated before the separation of Am. Also a cerium separation step would be required. Other difficulty is the oxidation of Am because the oxidation states IV and VI are unstable and must be stabilised with a strong oxidation reagent. The SESAME process, developed by CEA, uses an electrochemical method to oxidise the Am and then separate from Cm [97Adn].

An overview of recovery yields by the above mentioned PUREX processes is shown in Table 4.1

Other process developed by JAERI (Japan) separates the elements of HLLW into four groups (Four-Group Separation Process):

- All TRU elements including Np (V) are extracted with DIDPA after to reduce the nitric acid concentration to 0.5 M.
- Tc and platinum group metals are separated by precipitation through denitration.
- Sr and Cs are separated by adsorption with inorganic ion exchangers.

After the separation of TRU by DIDPA then are back-extracted with different reagents. Am, Cm and lanthanides are extracted again with DIDPA and then Am and Cm are back-extracted leaving lanthanides in DIDPA solvent.

Table 4.1: Recovery yields for PUREX hydrochemical reprocessing

Elements		Standard	Improved	Extended
U		99.88 %	99.9 %	99.9 %
Pu		99.88 %	99.9 %	99.9 %
Minor Actinides	Np	60 %	95-99.9 %	99.9 %
	Am			99.9 %
	Cm			99.9 %

Some long lived fission products such as ^{129}I, ^{99}Tc could also be separated within an improved PUREX process. Special off-line separation techniques are therefore necessary to separate Am and Cm and possibly Cs and Sr from liquid HLW.

The following flow sheet gives an overview on an advanced reprocessing of spent fuel based on hydrochemical processes.

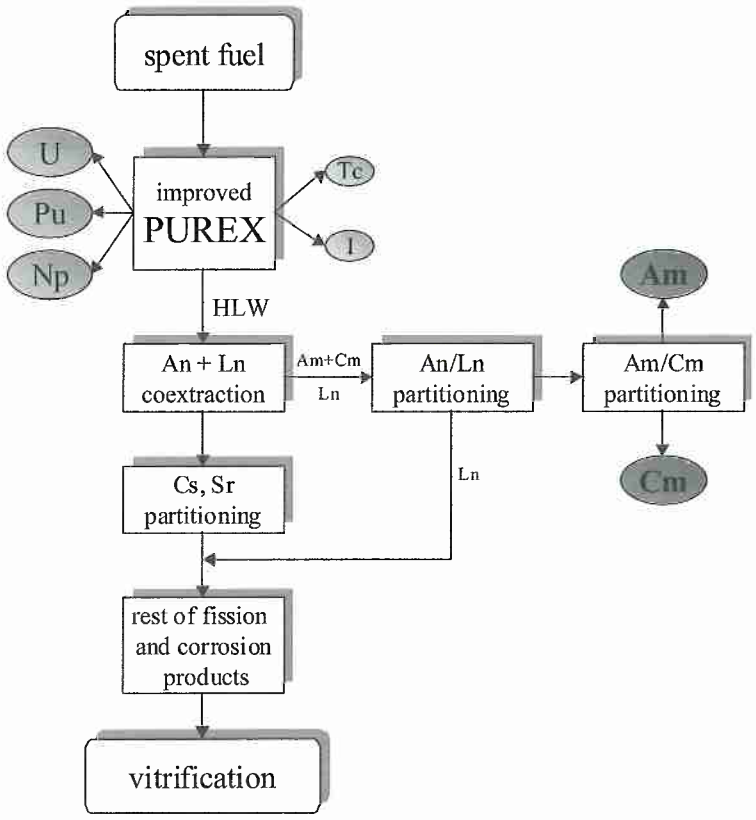

Fig. 4.1 Advanced reprocessing of spent fuel including MA recovery and separation of some long-lived radionuclides.

4.1.2 Possible application of hydrochemical processes to advanced fuels for transmutation

The possible use of hydrochemical processes on the reprocessing of the most relevant fuel forms for an ADS-type transmuter are summarised below.

- Metal fuels. The possible presence of a non-fissile metal, like zirconium, in the TRU alloy makes its dissolution very difficult in nitric acid and therefore the use of PUREX process.
- Oxide fuels. The mixed TRU oxides are similar to MOX, and therefore the PUREX process could be applied. However, the dissolution by nitric acid could have limitations due to the inventory of TRU, mainly Pu, and the presence of any inert matrix. For example, it has been shown that fuel with high Pu-content cannot be achieved in the existing reprocessing facilities since it does not dissolve quantitatively in the standard nitric acid solution (6 mol dm^{-3}). The dissolution behaviour of $(U_{1-x}Pu_x)O_2$ solid solutions in HNO_3 is characterised by a transition from easily solubility at Pu concentrations $x \leq 0.30$ to virtual insolubility at $x \geq 0.40$. Complete dissolution is perhaps achievable by addition of strong complexant such as fluoride or strong oxidants (e.g. Ce^{4+}, Ag^{2+}).
- Nitride fuels. Given that they are easily dissolved by nitric acid, the PUREX process could be applied on the reprocessing of these fuels. The recovery of N-15 from the off-gas dissolution stream could be made.
- Carbide fuels. These materials are easily dissolved in nitric acid but the formation of different organic compounds during the dissolution in nitric acid makes the use of the PUREX process difficult. A possible solution is the dry conversion of the carbides to carbonitrides or nitrides, which can be dissolved without difficulty.
- Composite fuels. The application of hydrochemical processes on the reprocessing of these fuels (ceramic-ceramic, ceramic-metal and metal-metal) can not be considered due to the difficulties in the fuel dissolution with nitric acid due to the inherent high actinide content of the actinide phase.
- Coated particle fuels. The same problems found on the dissolution of carbide fuels will invalidate the use of hydrochemical processes for the reprocessing of coated particles. In addition, the fuel particles have to crushed first to destroy the SiC layer which is insoluble.

In this respect a general comment must be taken into account. The high burn-ups in the ADS transmutation systems should increase the cooling time of irradiated fuel before reprocessing to avoid the high radiolysis effect on organic compounds that are proposed for the hydrochemical processes.

4.2 Pyrochemical processing

4.2.1 Introduction

For the recovery of radiotoxic minor actinides from irradiated spent fuel or from actinide targets following their transmutation, pyrochemical processes represent a promising alternative to hydrometallurgical separation methods. "Pyrochemical processes" are individual or combined operations by which materials are produced and refined in the absence of water (in contrast to "hydrochemical processes") at high temperatures. Such processes are widely used in industrial applications, notably for metals and alloys.

Already in the very beginning of the development of the nuclear industry, a number of pyrochemical processes have been investigated to separate the actinides from spent fuel. Commencing in the early 1950´s and extending through the 1960´s research and development work on pyrochemical and pyrometallurgical processes were conducted at various sites mainly in the US. The processes studied may be grouped into the following general categories: melt refining, zone melting, electrorefining, vacuum distillation, fractional crystallization, gas-solid reactions, and liquid-liquid extraction using either non miscible molten metal phases or non-miscible molten salt-metal phases.

During the 90´s renewed impetus for further investigation of these processes was due above all to progress and new concepts for the transmutation of fission products and actinides and for the development of the corresponding fuel cycles. New matrices are being designed for the elements to be transmuted, including heterogeneous recycling targets as well as dedicated fuels; longer-term perspectives also include new fuels for homogenous recycling concepts. Most of the scenarios considered require multiple recycling of targets or fuel to obtain high transmutation rates and thus a sufficient reduction in radiotoxicity. The far future options, such as dedicated fuel cycles and transmutation in molten salt reactors would certainly provide further impulse for the exploration of these processes. This is obvious in the case of molten salt reactors, for which pyrochemical techniques are the obvious and perhaps the only possible reprocessing solution.

For the reprocessing of nuclear fuel (targets) the following basic steps have to be considered:

- Separation of the fuel from the cladding material by mechanical techniques or
- Dissolution together with the fuel in molten salt at temperatures of the order of 500-800°C. Fluorides and chlorides are mainly proposed for this purpose although other media (phosphates, borates, nitrates, etc.) could also be considered.
- Separation of the actinides by electrorefining, transfer to a non-miscible liquid metal phase or selective precipitation. Partitioning by extraction is based on the differences in the thermodynamic stability of compounds or complexes of these elements and on changes in their oxidation state. Extraction by a liquid metal involves stripping by means of reducing agent. A multistage process improves, of course, the degree of refining and/or the recovery rate. In the

case of electrowinning or selective precipitation, the elements are recovered by a physical method.

The main objectives of these operations are of course very similar to those of hydrometallurgical processes:

- Selective extraction of the long-lived fission products (LLFP) and/or the actinides with high efficiency for subsequent recycling; the extracted product must therefore meet specifications for purity with respect to the remaining not extracted products.
- Minimisation of losses of the relevant elements in the various process steps
- Generation of process waste (not extracted elements in salt or metallic form and technological waste) compatible with a suitable treatment process.

The various concepts for pyrochemical reprocessing techniques that have been studied, developed and tested (in some cases at a pilot scale) throughout the world are summarised in Appendix B. A distinction must be made between the metal electrorefining as developed in US and Japan and the oxide electrorefining mainly developed in Russia. The metal fuel can be dissolved directly in the salt and deposition of the actinides is done on solid or liquid cathodes. This development process is well advanced and in the US an engineering scale facility was build for metal electrorefining of EBR-II fuel to recover uranium. About 1.5 t fuel was processed. In Japan the separation of plutonium (and americium) from uranium was demonstrated at laboratory scale and it was concluded that, from engineering point of view, more research is needed to clean the products deposited on the cathodes. In the oxide electrorefining a front-end conversion is normally foreseen, but in Russia the possibility of direct electrorefining was demonstrated. Also the oxide electrorefining process has reached the engineering stage for uranium and plutonium separation. For minor actinide recovery, however, the method still has to be explored.

4.2.2 Advantages and Drawbacks of Pyrochemical Processes

The reason for the selection of pyrochemical techniques in P&T concepts is based on a number of advantages:

- Good fuel and target solubility in molten salts, suitability for the treatment of highly refractory materials (e.g. ceramic-based targets). The dry reprocessing technique can be applied as well for the recycling of the homogeneously-distributed minor actinides in a fast reactor fuel as in the accelerator driven systems where the minor actinides are irradiated as oxides, carbides, metals or in inert matrices with concentrations of up to 20%.
- Possibilities to treat materials with minimum cooling time. Due to the higher radiation resistance of the inorganic reactants used in the processes, the reprocessing of short-cooled spent fuel is possible. The doubling time of the bred fissile material decreases (because the out-of-pile time of the fuel is reduced). For the spent fuel, cooling times as short as a few months seem possible, compared to the present five years and longer needed for aqueous reprocessing.

- Integration of reactor and reprocessing facility on one site (collocation). This is possible due to the compact process equipment. It would reduce the number of cost-intensive and complicated (national and international) transports of nuclear materials considerably.
- Lower criticality hazard due to the absence of water and thus of a neutron moderator in the process.
- Proliferation resistant process. Compared with aqueous methods, dry reprocessing of fuels results in less pure and thus more proliferation resistant fractions of Pu, Np or Am.

The pyrochemical processes have however also a number of drawbacks:

- Low decontamination factors compared with hydrochemical processes. It is difficult to separate individual transuranic elements, and there is generally always a certain degree of contamination with lanthanide fission product elements. If the recycled fuel is destined for a fast-spectrum reactor application, this contamination is usually of little consequence.
- Aggressive process media (molten salts and liquid metals), high melting points and thus high operation temperatures require very resistant materials.
- In the case of metallic fuels, a highly pure atmosphere and thus sophisticated technology is required.
- Processes difficult to develop for continuous operation (especially for electrowinning).
- Generation of neutrons when fluoride media are used.
- Technological and process waste needs special treatment.

4.2.3 Head- and back-end

It is evident that pyrochemical processes include not only a "core" process, in which the recyclable elements are extracted but also numerous ancillary operations at the front- or the tail-end of the main process:

- Prior preparation of the feed stream to ensure compatibility with the core process, e.g. decladding of solid fuel, halogenation of oxide fuel and dissolution in molten salts,
- Purification of the output stream, with recycling of contaminants back into the core process and treatment of the process and conditioning of waste materials in accordance with their chemical and radiological properties (e.g. vitrification),
- Decontamination of technological waste (e.g. crucibles) with recycling of contaminants back into the core process and conditioning of the actual waste materials (e.g. melting of scrap metal),
- Decontamination of all process off-gases, with recycling of contaminants back into the core process.

The performance characteristics required for the core process and for the ancillary operations depend directly on the losses of recyclable elements into the waste stream and on the desired degree of purity of the recycled elements.

If voloxidation of the spent oxide fuel is included in the pre-treatment, several fission products are distilled off including ^{129}I, which could be collected with 99.9 % yield, alkali metals, and some Tc and Ru. During the electro-refining process, 95% of the ^{99}Tc is collected together with the noble metal fission products in the bottom Cd layer (if used) of the electrolysis cell or in the anode baskets. For the transmutation of the two fission products, special targets have to be fabricated.

The waste produced throughout the process is expected to be minimal, since most of the materials (such as the eutectic salt mixture, the cadmium and bismuth metal and also the chlorine gas used for the chlorination) will be recycled. The LiCl-KCl mixture can be recycled after purification, while the salts of fission products are electrolytically decomposed and converted to oxides for vitrification in borosilicate glass. Non-recyclable waste will be solidified directly into a synthetic rock such as zeolite or sodalite with high mechanical integrity and leach resistance.

4.2.4 Possible application of pyrochemical processes to advanced fuels for transmutation

The possible use of pyrochemical processes for the reprocessing of the most relevant fuel forms for an ADS-type transmuter are summarised below.

- Metal fuels can be reprocessed easily by pyrochemical processes.
- Oxide fuels. The pyrochemical reprocessing of mixed TRU oxide fuels involves the conversion of the oxides to metal or chlorides, which can be dissolved in the molten salt electrolyte. An alternative is direct electrolysis of the metals from the oxide fuel in a salt electrolyte.
- Nitride fuels are highly compatible with the pyrochemical processes. The nitride can be dissolved in the anode and ^{15}N can be recovered from the off-gas.
- Carbide fuels. Like the oxide fuels, carbide fuels must be converted to metal or chlorides for electrorefining. Alternative pyrochemical methods for carbide fuel are also under development and fluoride processes seem encouraging.
- Composite fuels. The application of pyrochemical processes on the reprocessing of these fuels (ceramic-ceramic, ceramic-metal and metal-metal) depends strongly on the matrix and the actinide phase. Metals and nitrides can be reprocessed easily, oxides may require a conversion step as noted above.
- Coated particle fuels. The pyrochemical reprocessing of coated particle fuels will depend on the fuel design. The HTR-type coated fuel particle, which have multiple carbide coatings, are not easily reprocessed and a dedicated process to physically or chemically separate the fuel kernel from the coating is needed. Depending upon the process used for removal of the coatings,the oxide kernel can then be treated by a volatility process or an electrochemical process; the latter may involve the conversion of the oxide to the metallic state.

4.3 Summary

Pyrochemical processes offer more possibilities to reprocess MA fuel and targets than the hydrochemical processes. Especially for oxides (mixed transuranium, inert matrix or composite) and metals pyrochemistry is to be preferred. Nitride fuels can be processed by both methods without problems. In addition, the radiation stability of the salt in the pyrochemical process compared to the organic solvent in the hydrochemical process offers an important advantage when dealing with highly active spent MA fuel.

5. STATUS OF THE NUCLEAR INFRASTRUCTURE IN THE EU

5.1 Fuel/target fabrication

There is a limited possibility in Europe to handle and fabricate fuels/targets containing significant amounts of minor actinides such as americium and curium. Two civil facilities that can be used for this purpose have been identified: Atalante in France and the Institute for Transuranium Elements of the European Commission. They will be described below. In addition facilities exists in Russia and Japan, which might be of use to the ADS development programme.

In addition to the availability of facilities to fabricate fuels, other aspects need to be considered:

- The availability of material: At present americium and curium isotopes are available in small quantities (grams or tens of grams).
- Waste treatment: The treatment of storage of waste containing americium or curium is far from trivial, especially in case of liquid waste. Moreover, it will be very expensive.
- Transport of fuels/targets: Transport of fabricated fuel pins containing MAs is more complex as the neutron dose will be high. Extra shielding is needed.

5.1.1 Atalante (CEA, Marcoule)

The Atalante facility has developed a set of tools to manufacture americium-based ceramics. These tools may be used for other actinides also. They are installed in hot cells (HC) or glove boxes (GB) depending on the irradiation of the materials used. The steps of the fabrication process are the following:

- Chemical purification: The dissolution and the chemical purification of americium are performed in hot cells (C9) by established processes (chromatography, etc.).
- Elaboration and characterisation of powders: The precipitation and the thermal treatments on the powders are performed in a hot cell (C9). The characterisation is performed in glove boxes (L7) : grain size by Laser granulometry), specific area by BET.
- A laser granulometer and a BET will be implemented in a hot cell (C10) in 2001.
- Pellet fabrication: The whole pellet fabrication is available in hot cells (L6) or glove boxes (L7). The mixing of the powders is performed in small equipment, enabling the use of small quantities of materials (not exceeding 50 g). The mixture is compacted into pellets, which are then pulverized through a particle sieve in a low-retention granulator. A new type of press has been used that uses concentric plunger and die actuators. The pellets are sintered into a furnace with a capacity of 27 pellets at an operating temperature of about 1520°C with natural convection cooling. The sintering may be performed under controlled conditions in either oxidising or reducing atmosphere.
- Pellet characterisation: Visual and dimensional examination, and actinide content measurement by the following techniques (gamma spectrometry on a whole pellet, calorimetry

on a whole pellet, aliquots from a dissolution), ceramographic examination, X-ray diffraction analysis, chemical impurities measurement. The sintering characteristics of the products may be measured by a dilatometer (in a glove box).

- Pin elaboration: The pellets are placed into a fuel pin that is then pressured under helium and welded. The welding may be performed by laser or TIG. This system is available either in glove box and hot cells.
- Quality control: A quality assurance system according to ISO 9002 requirements is used for all the steps of this elaboration.

5.1.2 MA-laboratory (EU, JRC, Karlsruhe)

The Minor Actinide laboratory at the Institute for Transuranium Elements consists of two glove-box chains that are being build to fabricate fuels and targets containing significant quantities of minor actinides such as americium and curium. The main chain consists of 7 glove boxes that are shielded by a steel wall containing 50 cm water (neutron radiation) and 5 cm lead (gamma radiation). The limiting masses are 150 gram of ^{241}Am or 5 gram of ^{244}Cm. The processes in the glove boxes are partly automated by the use of robots and remote control, but actions are also carried out using telemanipulators.

The reference fabrication processes for are based on dust-free techniques such as SOL-GEL and infiltration; powder blending is excluded. These processes can be applied to fabricate mixed oxides, nitrides and composites. The installation is equipped with an infiltration device for powder preparation, calcination furnace, a bi-axial press, a sinter furnace (1650 °C) for reducing, inert and oxidising conditions, pellet inspection and metrology, pin filling and pin welding (TIG welding) of fuel pins up to 100 cm, and fuel pin characterisation (X-ray).

In addition to the seven glove boxes that form the core of the MA-lab, a separate chain of three glove boxes for preparation of powders containing americium or dirty plutonium by SOL-GEL technique will be installed. The shielding of these glove boxes, which are also operated with manipulators, is much less: 5 mm lead and 20 cm polyethylene. The limiting mass is 50 grams of ^{241}Am. The powder produced in this chain can be transferred to the main MA-lab chain where it can be processed further. Quality control is done according to ISO-9001.

The main chain of the MA-lab is expected to become operational in 2001, the second chain in 2002.

5.2 Irradiation facilities

A summary of the technical possibilities in the various reactors for fuel testing in the EU is given in Appendix C. It is clear that these reactors are well equipped to test fuels, in some cases also for tests with lead cooling under static conditions. However, the actual agreement on the possibility of

testing MA fuels or targets will depend on the safety reports and evaluation by reactor safety committees and cannot be guaranteed in advance. Also uncertainty exists about the possibilities for accepting and loading of MA-containing fuel pins as loading is often done in an unshielded or lead-shielded environment.

The reactors listed in Appendix B are all materials testing reactors with (epi)thermal neutron spectra. In most cases the neutron spectrum can be tailored to obtain a predominantly fast spectrum. Fast reactors for fuel test will not be available in the EU after 2004, the planned date for the operation of Phénix.

5.3 Post-irradiation examinations

The review of the European hot laboratories, given in Appendix D, shows that no specific regulations pertain to the acceptance of irradiated targets containing minor actinides. In general, it is quoted that the presence of trace amounts of the minor actinides "as present in irradiated nuclear fuel" is accepted in the fuel hot cells. Limits on the amount of fuel that can be present in the hot-cells are based on gamma dose rate and potential criticality.

Let us first consider the order of magnitude of the quantity of minor actinides present in irradiated nuclear fuel. As an example, we assume that ten full length fuel rods are allowed in the NDT (non-destructive testing) hot cell, which roughly corresponds to 25 kg of irradiated fuel. The concentration of minor actinides in irradiated fuel of a burnup of 55 GWd/tHM and a cooling time of one year, is given in Table 5.1. In one kilogram of irradiated UO_2, one has about 0.25g of Am and 0.1 g of Cm. In the case of MOX fuel, the amounts of both Am and Cm are significantly higher: about 1.8g of Am and 1 g of Cm, again per kilogram of fuel. This means that according to the current regulations, one accepts 6 g of Am and 2.5 g of Cm in the case of UO_2 rods and up to 45 g of Am and 25 g of Cm dispersed in ten irradiated MOX fuel rods. While the dose rate and contamination risk may be acceptable when the minor actinides are embedded in the irradiated fuel rods, this will not automatically be the case for targets containing elevated concentrations of the concerned MA's.

The beta/gamma shielding of hot-cells that are currently in use for research on nuclear fuel obviously is sufficient regarding the gamma activity of targets containing high concentrations of the minor actinides. The leak tightness of the shielding may be problematic for some hot laboratories, especially with regard to the acceptance of targets that are damaged. When calculating the radiotoxicity of Pu, Am and Cm on the basis of the isotopic composition given in Table 5. 1, and expressing the annual limit of intake, ALI, (ICRP-68) as a function of weight, one finds that the ALI of Americium is of the same order as that of Plutonium while for Curium the ALI is two orders of magnitude lower. In view of the high radiotoxicity of the considered isotopes, it will be imperative to have leak-tight hot-cells in which irradiated targets containing high amounts

of Am and Cm are to be handled and/or investigated. There are, at present, several hot-laboratories that do have glove boxes inside their hot-cells. Regarding the shielding for neutrons, a more elaborate study should be carried out. At present, there is no specific neutron shielding of the hot-cells that are currently used for the investigation of irradiated fuel.

Table 5.1: Concentration of the minor actinides in irradiated UO_2 and MOX fuel at BU of 54 and 56 GWd/tHM respectively, one year after end-of-burnup (EOB +1) (data calculated on the basis of experimental results obtained in the framework of the ARIANE project).

Actinide	UO_2 (wt%)	MOX (wt%)	ALI (intake) (g)	ALI (inhalation) (g)
237Np	6.24E-02	1.27E-02		
Np (all)	**0.062**	**0.013**	**7.E-03**	**4.E-05**
238Pu	2.74E-02	1.00E-01		
239Pu	3.42E-01	5.43E-01		
240Pu	2.39E-01	1.00E+00		
241Pu	9.63E-02	3.97E-01		
242Pu	7.53E-02	5.80E-01		
244Pu	7.05E-06	1.00E-04		
Pu (all)	**0.780**	**2.62**	**2.E-06**	**1.E-08**
241Am	8.30E-03	4.53E-02		
242Am	6.11E-05	5.90E-04		
243Am	1.52E-02	1.34E-01		
Am (all)	**0.024**	**0.180**	**2.E-06**	**1.E-08**
242Cm	1.87E-03	3.28E-03		
243Cm	4.68E-05	5.93E-04		
244Cm	6.46E-03	9.33E-02		
245Cm	2.95E-04	6.49E-03		
246Cm	6.52E-05	1.62E-03		
Cm (all)	**0.009**	**0.105**	**3.E-08**	**1.E-10**

The following conclusions on the status of the European infrastructure can be drawn:

- There are no specific problems to be expected for the investigations of *intact targets* with *low concentration of neutron emitters*. The required gamma shielding for the investigation of irradiated MA targets is not different from the shielding necessary to investigate irradiated fuel pins. The situation is changed when the targets might either be defective or when the investigations are not limited to NDT alone.
- In view of their high specific radiotoxicity, investigations on open sources of MA targets are to be conducted inside leak-tight glove boxes. Although not every laboratory has its full chain of cells equipped with inner glove boxes, this is the case for some hot labs and most of the hot

labs have at least one cell equipped with a glove box. Past experience with Pu containing fuels is relevant to this aspect of the investigations.

- When MA targets with elevated concentrations of Cm are to be investigated, one should specifically look at the problem of neutron dose, as the current hot cells are not specifically designed for high neutron-emitting samples. It seems logic that a full assessment of the acceptance limits on irradiated MA containing targets should be made in the near future. One should then look at the entire flowchart of the examinations to be carried out and the type of work that is to be performed in each of the utilized cells.

5.4 Reprocessing

Dedicated research facilities for hydrochemical reprocessing are available in France (CEA, Marcoule) and U.K. (BNFL, Sellafield) where extensive studies are being performed on spent fuel material. Facilities for smaller scale hot-test are available in other countries and the JRC-ITU (Karlsruhe, Germany). In addition, European Universities are strongly involved in this research through the Framework Programmes, especially in the fields of synthesis and molecular modelling of new extractants.

The realisation of an infrastructure for studies on pyrochemical reprocessing is on-going in Europe at present. In several laboratories facilities are being installed for electrorefining of active materials (irradiated and unirradiated): AEAT (Harwell, UK), BNFL (Sellafield, UK), CEA (Marcoule, France) and JRC-ITU (Karlsruhe, Germany). CIEMAT and ENEA are constructing support laboratories, where work with inactive materials (e.g. SIMFUELS), Th and U can be performed. Many of the studies rely on long-term experience in universities (e.g. Grenoble, Toulouse, Valladolid etc.) and on international collaborations with Japan or US.

5.5 Databases

Databases relevant to the subject advanced fuel cycles can be categorised as follows:

1. Fuel material properties: Many databases on properties of materials exist and some of them contain information about relevant actinide compounds. Examples are the NEA Thermochemical database Project [95Sil,2001Lem], the INSC Material Property database of ANL or the IVTAN Thermochemical database of the Russian Academy of Sciences. However, with respect to the compounds of transuranium elements these databases are incomplete, as limited experimental data for these compounds exists. Also the scope of most of the databases is not on fuel technology and as a result not all relevant properties are included. Because of these reasons, ITU has started an activity on an actinide/lanthanide database [2001ITU].
2. Irradiation data: The irradiation data on advanced fuel forms have not been collected systematically in databases.

3. Fundamental data for pyrochemistry: Data on Gibbs Energies of formation of actinide, lanthanide and fission product chlorides are available in several databases. In this respect it should be mentioned that the actinide chlorides (U, Np, Pu, Am) have been carefully evaluated for the NEA-TDP. An assessment of the lanthanide trichlorides is also available [2001ITU].
4. Separation data: There is a significant amount of experimentally determined distribution coefficients for pyrochemical separation in chloride salts. However, they have not been collected systematically in databases.

5.6 Summary

There still exists a good infrastructure in Europe for research on fuel and fuel processing which covers the whole range from fuel fabrication to fuel reprocessing. However, the availability of these facilities is not guaranteed for the period up to the start of the XADS.

For the fuel fabrication two laboratories can handle larg(er) amounts of minor actinides. This seems not sufficient for the development of MA fuel pins for ADS. For the irradiation, a number of research reactors is available. However, no fast spectrum reactor is expected to be in operation after 2004 (expected closure of Phenix). The XADS is therefore an important tool for the fuel development of future ADSs. Hot cells for post-irradiation examinations of nuclear fuels are available in several EU countries. However, only a few can accept irradiated MA fuel at present.

For reprocessing studies dedicated facilities exists in the EU, though the number of hot pyrochemical facilities is limited. At AEAT, BNFL, ITU and CEA such facilities are under construction at present.

6. CONCLUSIONS

6.1 Fuel forms

The state-of-the-art of the technology of the most relevant fuel forms for an ADS-type transmuter have been described in this report. The following conclusions have been reached for the individual fuel forms:

- Metal fuel has interesting thermal and mechanical properties and fuel designs to deal with the extensive swelling of this fuel form exist. Metal fuel based on U could be an interesting option but does not meet the criteria of this study. A mixed transuranium metal fuel does not seem feasible (low melting temperature, limited mutual solubility). Zirconium seems to be to most likely candidate as non-fissile matrix but there is considerable uncertainty about the (mutual) solubility of neptunium and zirconium. In addition, considerable safety measures need to be implemented in the fabrication process due to the pyrophoric nature of metals and the high volatility of Am. In addition, the compatibility of metal fuel with liquid lead or lead/bismuth is not certain.
- Oxide fuels have the general advantage that they are stable in air, which makes the safety requirements for the fabrication process less demanding. Three types of homogeneous oxide fuel have been discussed, mixed transuranium oxide, inert-matrix mixed oxide and thorium-based mixed oxide fuel. Their fabrication is close to the current MOX fabrication technology. However, the operating temperature of (mixed) oxide fuel is the expected to be high, caused by the relatively poor thermal conductivity, which may lead to limits in the linear rating. When thorium oxide is used as matrix, this might be less significant, but the eventual drawback of ^{233}U formation has to be evaluated.
- Nitride fuel has good thermal conductivity and the actinide nitrides show a mutual miscibility in a wide range of compositions. Nitride fuel is expected to have a good behaviour during normal operating conditions. The relatively high dissociation pressures of PuN and AmN at elevated temperature form, however, the major disadvantage of the mixed transuranium nitride fuel during off-normal operating conditions in the case of severe accident (CDA). It requires very specific fuel and core design for safe operation nitride fuel. Nitrogen-15 must be used for fabrication in order to prevent a radiotoxic impact to the environment, which makes the fabrication process more demanding.
- Carbide fuels, in spite of their good thermal and mechanical properties, are considered of limited interest. Helium bonding was found to be sufficient to reach high burnup. Unlike the nitride fuel, there is no safety concern regarding the case of severe accident (CDA). The highly pyrophoric nature of the carbide compounds requires an inert atmosphere for fabrication. In case of transuranium carbide fuel, a strict control of stoichiometry is required in order to obtain a monocarbide phase. The compatibility with wet reprocessing is only given by transforming the spent fuel to a carbonitride. Overall, this fuel is considered less interesting than most other fuels described in this report.

- Composite fuels (ceramic-ceramic, ceramic-metal and metal-metal) have the advantage that the properties of the fuel can be tailored to a certain extent by a careful selection of materials. However, the number of relevant material combinations is limited, the most promising being MgO-$(Zr,An)O_{2-x}$ for ceramic-ceramic type, steel-$(Zr,An)O_{2-x}$ for ceramic-metal type and Zr-(An,Zr) for metal-metal type. It should be noted that the $(Zr,An)O_{2-x}$ oxide phase is favoured to the (An,Zr) metal phase. The fabrication of composite fuel is considerably more difficult, especially in case specific requirements for the size and distribution of the dispersed actinide phase. Thus the development costs of this type of fuel may be considerable, if not prohibitive, compared to the more simple homogeneous fuel. Limited experience exists in this field. Also the irradiation behaviour forms a major uncertainty, especially the effect of locally high production of helium and fission gases on the mechanical behaviour.
- Coated particle fuel is essentially a special form of composite fuel. It is well known from the HTR development and offers the possibility of gas retention. However, for a fast system a completely new concept needs to be developed, as carbon will act as a neutron moderator. The costs of this will be considerable.

In general, the present evaluation has made clear that significant experience exists for uranium-based fuel forms but that there is a lack of knowledge about the characteristics and fabrication technology for the corresponding uranium-free fuel forms. However, qualitatively it can be concluded that the physico-chemical properties of homogeneous uranium-free fuel forms are generally poorer than the corresponding uranium forms as relevant properties (melting point, thermal conductivity and chemical stability) decrease systematically for most compounds going from Th to Am. These poor(er) properties might be compensated by the choice of the matrix of the fuel, but only a limited number of adequate materials have been identified, none of them being ideal in all respects and compromises thus have to be found.

The present evaluation also made clear that almost no experimental information is available on the irradiation behaviour of uranium-free fuel forms. The EFTTRA-T4 experiment is the only known case in which minor actinide in a non-fertile matrix has been irradiated. This experiment has demonstrated the importance of the helium gas accumulation in the fuel, leading to intolerable swelling if no engineering measures are taken. This relatively large helium production in the fuel (at high burn-up combined with a large production of fission gases) is an unique feature of minor-actinide containing fuels/targets, and could br considered the *Achilles heel* of MA fuels. This will require a special design, as the gas production is at least one to two orders of magnitude higher than in standard fast-reactor mixed oxide fuel. As a result, the americium content of the fuel might be limited by design considerations.

The presence of Curium in the fuel will have a big impact on the fuel cycle, which is not yet sufficiently understood. Apart from the required increased radiological protection (against neutrons) the effect on fuel behaviour needs to be analysed in depth, especially the effect of alpha decay which is continuous (starting immediately after fabrication).

Because of the general lack of knowledge on the properties and behaviour of uranium-free fuels, it is not possible to reach firm conclusions on the best fuel forms for an ADS transmuter at this moment. However, the most promising areas on which the European research should be focused can be identified. It is evident that oxide and composite fuels, and to a lesser extent nitride fuels, are the most promising candidates for further research. Composite fuel is considered the most innovative of these because the properties can be tailored to the application. However, the limitations of this fuel type need to be investigated thoroughly. Mixed transuranium oxide fuel is the closest to current European MOX fabrication technology and for this reasons it is an important candidate to consider. It has clear advantages with respect to fabrication and thermo-mechanical stability but less promising thermal properties may limit its application. Mixed nitride fuel forms a reasonable alternative to the (homogeneous) mixed oxide if high temperature stability can be achieved, for example by addition of ZrN.

6.2 Fission product targets

Transmutation of three fission products is considered:

- Technetium metal is an excellent target material for transmutation of ^{99}Tc.
- Sodium iodide (NaI) is considered to be an acceptable target material of ^{129}I though its properties are mediocre. More details about the positioning and the thermal environment of the targets in the ADS (core, core periphery, lead) need to be known to make a more detailed analysis.
- Caesium transmutation does not seem feasible.

6.3 Fuel processing

The partitioning of minor actinides is still subject of research. Hydrochemical and pyrochemical techniques are under investigation. Both have been considered in the frame of this report and the following conclusions have been reached:

- Hydrochemical processes have a high potential to separate the minor actinides from spent fuels as demonstrated by extensive research, especially in Europe. However, the major drawbacks of this technique for uranium-free fuels for ADS are:
  - The limited solubility of many fuel forms considered.
  - The limited stability of the organic extraction molecules in high radiation fields expected for spent ADS fuels/targets.

  As a result, the hydrochemical processes are considered to be more relevant to the first stratum reprocessing of LWR or FNR fuel than for the second stratum reprocessing of ADS fuel/targets
- Pyrochemical techniques seem to offer the highest potential for reprocessing of ADS fuels/targets due its good compatibility with most fuel forms and its high radiation resistance.

In addition, the compactness of the technique is an important advantage. Electrorefining is generally considered to be the most promising pyrochemical method and it is being investigated world-wide, especially in US, Japan and Russia.

6.4 Infrastructure in EU

There still exists a good infrastructure in Europe for research on fuel and fuel processing which covers the whole range from fuel fabrication to fuel reprocessing. However, the availability of these facilities is not guaranteed for the period up to the start of the XADS.

- The European infrastructure for research on fuel fabrication and processing is becoming limited. Only two laboratories are able to fabricate MA-based fuels: Atalante in Marcoule (CEA) and ITU in Karlsruhe (EC-JRC).
- For irradiation experiments and tests, a number of research reactors is available. However, no fast spectrum reactor is expected to be in operation after 2004 (planned closure of Phénix). The XADS is therefore an important tool for the fuel development of future ADSs. Hot cells for post-irradiation examinations of nuclear fuels are available in several EU countries. However, only a few can accept irradiated MA fuel at present.
- For reprocessing studies dedicated facilities exists in the EU, though the number of hot pyrochemical facilities is limited. At AEAT, BNFL, ITU and CEA such facilities are under construction at present.

7. RECOMMENDATIONS

7.1 Fuel research

In order to test an advanced fuel for transmutation of the transuranium actinides in the XADS, a careful planning of the development phase is required because many fuel forms are still investigated in parallel and no fuel pin design is yet available for any of these forms, as demonstrated in the preceding chapters. It is also known from past experience that fuel development is very time consuming because long irradiation tests are involved. To test fuel pins or fuel assemblies of advanced fuel forms in the XADS around 2020, a carefully co-ordinated European effort is required, for which the following suggestions are made:

1. A specific irradiation experiment in combination with a dedicated out-of-pile programme on the determination of the basic properties of mixed oxide fuels (with and without matrix) and a in-depth safety analysis should establish the limits of the use of this fuel form for ADS. The activities should aim at providing the following information:
   - Thermal properties of $(Th,Pu,MA)O_{2-x}$, $(Zr,Pu,MA)O_{2-x}$ and $(Pu,MA)O_{2-x}$ (thermal conductivity and melting point as a function of composition and oxygen content; oxygen potential of the actinide phase).
   - The vapour pressures of the actinide metals over the oxide phase.
   - The irradiation behaviour of $(Th,Pu,MA)O_{2-x}$, $(Zr,Pu,MA)O_{2-x}$ and $(Pu,MA)O_{2-x}$.
   - Compatibility with liquid metal coolant and cladding material.
   - The expected behaviour of mixed oxide fuels during off-normal conditions.

   A period of 4-5 years is required for the out-of-pile part, provided the necessary infrastructure is available (see 7.4). For irradiation tests, a longer period (8-10 years) would be required. The ongoing activities of the EFTTRA collaboration and of CEA (ECRIX and CAMIX experiments) should be integrated in this.

2. The research on composite fuels for ADS transmuters should be focussed on the MgO-$(Zr,An)O_{2-x}$ ceramic-ceramic and the steel-$(Zr,An)O_{2-x}$ ceramic-metal fuels. Many aspects of these fuels need to be investigated in detail such as fabrication, irradiation behaviour and fundamental properties. The following aspects are considered critical for the applicability of these fuel types:
   - Phase relationships in the complex multi-component systems need to be established to exclude interaction or low-temperature eutectic melting, for which some evidence in case of the ceramic-ceramic fuel has been presented.
   - The optimum size and form of the dispersed phase need to be established, both from theoretical and practical (fabrication) point of view.
   - The irradiation behaviour of these fuel types, especially with respect to the effect of local accumulation of helium and fission gases in and around the dispersed phase.

- The behaviour during accident conditions, especially the possible effects of phase separation in composite materials.

A period of 6-10 years is required for a systematic research programme on these topics. It is of key importance that the first screening tests (in-pile as well as out-of-pile) are started as soon as possible.

3. Nitride fuel should be considered as back-up solution. Because MA-containing nitride fuel is studied extensively for the ADS concepts in Japan and Russia, it should not become a major topic of the European research. It is recommended to seek a collaboration with the Japanese and Russian activities in this field. The CONFIRM project in the 5th Framework Programme of the EU (irradiation of (Zr,Pu)N fuel) should be starting point for such a collaboration.

4. In about 10 years, the primary fuel form for the European ADS development could be selected on the basis of the experimental results of the proposed activities. However, it should be clear that more details about the system design of the ADS must be fixed rapidly. Especially the type of cooling has a big impact on the fuel: operating temperatures in gas-cooled cores will be higher than in liquid-metal cooled cores, and the possible corrosion by the coolant needs to be considered in liquid-metal cooled cores. Also the safety approach has an impact of the fuel selection and fuel design, and different scenarios need to be considered.

7.2 Fission product targets

5. The research on targets for iodine transmutation should be continued, taking into account the specific conditions in the ADS core. No clear incentive exists for the study of targets for transmutation of caesium.

7.3 Fuel processing

6. The research activities on pyrochemical reprocessing in Europe should be intensified. Its use for the processing of mixed transuranium oxide fuel and composite fuels, which have been identified in this report as the main candidate fuel forms, has to be demonstrated. It is important in this respect that additional facilities will become available.

7.4 European infrastructure

7. There are only two fabrication laboratories in Europe able to handle americium and curium for R&D on fuels and fuel processing: ITU (JRC, Karlsruhe) and Atalante (CEA, Marcoule). Taking into account the volume of basic data needed to demonstrate the feasibility of dedicated fuels for ADS in a lot of fields, it is obvious that both laboratories at least are needed to manage a complete programme over a period of 10 to 15 years. Complementary actions have to be taken between the two laboratories to cover such a programme. This is an additional argument to focus in Europe the R&D on MA-based oxide fuels (homogenous or composite fuels), as MA-based nitride fuels have been already investigating at JAERI, Japan for about 10 years and metallic fuels have been recently proposed by the American as a promising candidate for the ATW.

8. For the testing of the irradiation behaviour of fuels and targets for transmutation in ADS, the XADS is an important instrument as no fast reactor will be available in the EU after 2004. It is felt that the XADS must be integrated with a fuel fabrication facility and hot cells at one site to avoid time-consuming transports and enhance the efficiency of the research.

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Appendix A: The Mandate of the FFP Group (by H. Aït Abderrahim, G. Heusener, M. Salvatores)

The mandate of the subgroup (FFP) should cover the following three areas and should produce in the next 4 months a state of the art report and recommendations.

I Fuel for the ASAP Demonstrator.

The subgroup is to analyse the fuel situation for the ASAP Demonstrator. The following items should be investigated:

- Existing fuel available and usable? (SNR 300, SPX) – Compatibility with different coolants.
- Which modifications of the existing fuel are needed? Costs? Who can do the modification? (Example: There is probably quite a bit of Am 241 in the SNR 300 fuel; can the SNR 300 fuel be handled in normal facilities or is additional shielding necessary)
- Additional R and D necessary?
- If additional R and D effort is needed, are appropriate activities underway?

II Dedicated Fuel for Burner Machines and LLFP targets

It is assumed, that the MAs (and possibly Pu) are burned in dedicated Transmuters; Fuel with high fractions of MAs is needed for these reactors. This kind of fuel does not exist today. Respective R and D programs are underway, though most probably the present effort is not sufficient. In the case of ADS burners, inert matrices probably can be used. Th based fuel also is a possibility. Oxides, Nitrides and Metal, but also carbides, are considered world-wide. The subgroup is asked to make a synthesis of all (European) activities, identify the specific problems of the dedicated fuel, e. g. high contents of Pu 238, high radiation levels, need for remote handling etc.

Analysis of available irradiation facilities, hot laboratories able to handle large amounts of MA's (both Am and Cm), type of post-irradiation examination as well as on-pile possibilities at the different hot cells and irradiation facilities, need for specific loops and irradiation conditions. The need for a co-ordinated program to establish a physico-chemical database should be evaluated. Recommendations for future activities, first attempt to have a global program together with the US and Japan?

Finally, Long-Lived Fission products (LLFP) targets for transmutation should also be considered.

III Fuel Processing

Dedicated fuels most probably need special processes for reprocessing and fabrication. The kind of fuel, way of reprocessing and fabrication are closely connected. The FFP subgroup is asked to put together a list of possibilities, identify the advantages and disadvantages, and describe the state of the art. Wet and dry processes shall be considered. Also in this field possible synergies should be indicated with respect to similar programs in the US, Japan (and possibly Russia).

Appendix B: Concepts for pyrochemical reprocessing technology

B1. USA

The American IFR concept based on metallic fuels is today the most mature of the pyrochemical processes and has been tested experimentally to recondition almost 1.5 MTHM of irradiated metallic driver fuel and blankets at ANL-West, Idaho. The basic principles of pyrochemical reprocessing in the Argonne National Laboratory (ANL) for the Integral Fast Reactor (IFR) concept were proposed and developed in the Argonne-East laboratories in Chicago. Glove-box studies on electrorefining of metallic fuel in molten chloride media under pure argon atmosphere began in the mid-1980s. Constant improvements of the process and the electrorefiner equipment were made.

Today in the fuel conditioning facility (FCF) at ANL-W, the fuel is anodically dissolved in an electrolyte of LiCl-KCl at about 500 °C; uranium is recovered on a solid cathode, and the transuranic elements and fission products remain in the electrolyte salt. The TRU content of the fuel and hence the salt is rather low, and the salt with all of its constituents is converted into a glass-ceramic composite waste form for disposal. An anodic dissolution test on EBR-II fuel showed that most of the alloy was dissolved into salt while a little cladding material remained as residue. Recovery of uranium on the solid cathode has been thoroughly demonstrated with 10 kg deposited in a dendrite form. The condition of effective deposition on the solid cathode was investigated by changing the ratio of cathode to anode surface area at CRIEPI. At present the work is mainly concentrated on the development of a large-capacity (150-350 kg/day) uranium electrorefiner with an anode/cathode system consisting of concentric cathode tubes that amass a deposit of uranium electrotransported from anode baskets rotating in the annuli between cathode tubes. Electrodeposited uranium is removed from the tube surfaces by knife-blade scrapers attached to the rotating anode baskets, and the uranium falls to the bottom of the electrorefiner where it is collected. This electrorefiner should allow reprocessing of several tons of metallic fuels stored in the US.

Argonne-East also envisages processing of spent PWR oxide fuel in identical electrorefiners. Oxide fuels to be treated by this process must first be reduced to metal. ANL experimentally evaluated several methods, such as salt transport, the Zn-Mg process, Mg extraction and lithium reduction. The lithium process has the advantages of a lower operating temperature, less corrosive environment and small amounts of salt waste. Single element tests used uranium, plutonium and other element demonstrates the reduction to metallic form in LiCl-KCl at above 500°C, resulting the formation of lithium oxide, which can be electrolytically decomposed for recycling. The results have shown that the actinide oxides are completely reduced if the Li_2O concentration (formed in the medium during reduction) does not exceed 3 wt% (N.B. CRIEPI found 5wt%).

B2. Russia

In Russia the Research Institute of Atomic Reactors (RIAR) in Dimitrovgrad has successfully demonstrated the technical feasibility of the pyro-electrochemical reprocessing of oxide fuel by direct electrorefining without reduction. Several kg of spent fuel from the BOR 60 fast breeder reactor were already reprocessed.

Russian experience with molten salt dates from the 1950´s with the creation of the High-Temperature Electrochemistry Institute at Ekaterinbourg. A wide range of laboratory studies (characterization of salt structures, electrochemistry, kinetics, etc.) was carried out there in molten chloride media on uranium, thorium and simulated fission products. Work began in 1964 on plutonium in the Electrochemical Processes Laboratory of the RIAR-Institute to specify a series of elementary operations for the fabrication and reprocessing of UO_2 and MOX fuels by the vibrocompaction method.

Research activities today (potentiometry, voltammetry, etc.) focus on the behaviour of neptunium and americium for the fabrication of oxide fuel containing these elements. In the so-called DOVITA-process developed in Dimitrovgrad, the oxide fuel is converted into chlorides. UO_2, PuO_2 as well as $(U,Pu,Np)O_2$ are separated by electrolysis in a melt of NaCl-KCl at 650°C. The transuranium elements are precipitated sequentially as the oxo-chlorides or oxides from the NaCl-KCl melt by gassing with Cl_2/O_2 and adding Na_2CO_3. Since lanthanides and the transplutonium elements (Am, Cm) have similar behaviour, a fractionated precipitation of the oxichlorides is proposed in order to obtain an Am, Cm fraction with a sufficiently low lanthanide content. From the technological point of view, this is however a cumbersome step; therefore it would be certainly preferable to develop an electrorefining process also for Am and Cm. The Arzamas and Kurchatov Institutes have also proposed a molten salt reactor concept for Pu and minor actinide management purposes.

B3. Japanese Research Concepts

Japan's research and development activities on pyro-electrochemistry are mainly carried out by:

- CRIEPI (Central Research Institute of Electric Power Industry) with the development of a metallic fuel process.
- TEPCO (Tokyo Electric Power Company), who has embarked recently in a large collaboration with RIAR to study reprocessing of MOX fuel.
- JAERI (Japan Atomic Energy Research Institute) who is investigating the potential of nitride fuels for minor actinide transmutation
- JNC (Japan Nuclear Fuel Cycle Development Institute) has started an evaluation of different pyrochemical reprocessing technologies in view of future nuclear fuel cycles in Japan.

B3.1 CRIEPI: Actinide recycling in combined FBR and LWR cycles

CRIEPI is investigating the technology of recycling actinides from metallic FBR or reduced oxide fuels, and also their recovery from high-level liquid waste. The processes are essentially based on the metal fuel cycle as previously described using molten chlorides (LiCl-KCl) in contact with liquid metal (Cd or Bi). This recycling system should be operated flexibly by combining LWR and FBR fuel cycles. Its main features are the electrochemical deposition of uranium and plutonium onto solid and liquid Cd cathodes and the recovery of transuranium elements from the salt by reductive extraction (Cd or Bi) or electrorefining. Whereas the US and Russian concepts focus mainly on the reprocessing of U and Pu, CRIEPI has performed extensive experimental work on actinide/lanthanide separation in molten chloride and liquid metal (Cd, Bi) media by electrodeposition or by liquid-liquid extraction. Laboratory scale experiments were performed on minor actinides in collaboration with several organisations, including the University of Missouri for glove box tests and the Transuranium Institute in Karlsruhe for hot-cell verification experiments on genuine metallic targets the latter started mid-1999.

Results have shown that the electro-refining of minor actinide metals in molten LiCl-KCl at 450 °C seems feasible even for Am. The alternative approach is the combination of electro-refining with a reductive multistage extraction from the molten salt into molten Cd or Bi. In these tests conducted at trace levels, promising recovery rates of 99.9% for Am were achieved.

In the larger installation set up at ITU, a demonstration of the technical feasibility of the electro-refining or reductive extraction concept is to be made. Furthermore conditions to improve the electro-refining of Am from lanthanides are being investigated. This installation will be the first to be operated at the gram-scale of minor actinides. As mentioned above, in the conditioning facility of the EBR-II fuel at Argonne National Laboratory only uranium is separated by electrorefining, the transuranium elements remain together with the fission products.

B3.2 CRIEPI: Pyrochemical separation of transuranium elements from high-level liquid waste

The process investigated at CRIEPI to recover transuranium elements from high-level PUREX wastes consists of the following steps: denitration to oxides, chlorination, reductive extraction and electrorefining in a LiCl-KCl/Cd or LiCl-KCl/Bi system. After denitration and leaching with water to remove the soluble alkali-metal nitrates, the undissolved oxides (mainly of actinides, rare earths and transition metals) and platinum-group metals are converted into chlorides in LiCl-KCl at temperatures >700 °C.

The mixed chlorides are reductively extracted or electrorefined in contact with liquid cadmium or bismuth. Although thermodynamic data for actinides and rare earth elements in this system are still needed to establish the separation process, small-scale experiments showed that already over

99% of each actinide could be recovered from simulated waste. Further technological assessment requires a pilot-scale demonstration with the full range of actinides.

B3.3 JAERI

Since the mid 1990´s, JAERI has been working on a fuel cycle combining nitride fuel with pyrochemical fabrication and reprocessing methods in molten chloride media at a laboratory scale. While encouraging preliminary results have been obtained with uranium, major R&D work remains to be done on the transuranium nuclides. In the double strata concept foreseen in the OMEGA program, JAERI investigates the possibility of using nitride fuels and pyrochemical processing in the second stratum. In this case the recycling of ^{15}N to avoid the formation of ^{14}C during irradiation is much easier. This proposal is supported by a major theoretical and experimental research program and the electrorefoning of NpN and PuN has already been studied at JAERI

B3.4 JNC

At the JNC the examination of pyrochemical processes is performed on the laboratory and on the engineering scale and aims at an evaluation of the potential of this technology from an economic point of view. Comparative assessments between dry processes and aqueous processes will be done in the near future at JNC in collaboration with utilities and companies.

A glove box facility for plutonium experiments with a capacity to treat 200 g of fissile material (MOX) is installed in Tokai-mura in collaboration with CRIEPI. Emphasis is mainly put on the performance and engineering aspects of the process. The Pu behaviour is studied in each step of the pyrometallurgical process, i.e. fuel reduction, electrorefining, liquid-liquid extraction, chloride distillation, metal distillation, oxidation. Emphasis is mainly put on the performance and the safety aspects of the process

B4. Europe

A network of six European research organisations has submitted an R&D proposal on pyrochemical reprocessing to the 5th Framework Program for Technological Research and Development of the European Commission. Ther partners are CEA (Marcoule, France), together with the ENSCP (Paris), the INPG (Grenoble) and the IUSTI (Marseille), CIEMAT (Madrid, Spain) ENFA (Casaccia, Italy), ITU/JRC (Karlsruhe, Europe), together with CRIEPI (Tokyo, Japan), NRI (Rez, Czechoslovakia) and BNFL (Sellafield, UK), together with AEA-T (Harwell, UK). This program should revive European research in the area of pyrochemical reprocessing and contribute to establish a long-term European expertise in this area.

The main objective of the proposal, which includes both conceptual studies and experimental work, is to provide basic data, especially on the minor actinides. Many aspects of the thermodynamics of

actinides in metal and salt media are insufficiently known. Based on the presently available data it is not possible to propose an operational process flowsheet suitable for these elements. The program therefore addresses the following issues:

- Experimental acquisition of basic data on elementary chemical processes, involved in the major process steps: fuel conversion to the halide form, radionuclide separation by electrolysis or metal/salt extraction, recycling of the process reactants.
- Liquid and solid process waste treatment;
- Identification and selection of materials compatible with the harsh conditions of pyrochemical processes (high temperature and corrosive media);
- Concept system studies: performance assessments, estimates of material flows and characterisation of waste produced by the different pyrochemical processes.

Experiments with radioactive materials will be carried out in facilities operated by the CEA (ATALANTE) and ITU. Supporting experiments will be performed on simulated fuel.

Appendix C: European Research Reactors

C1. The BR-2 Materials Test Reactor

The BR-2 is a high flux engineering test reactor that differs from comparable materials test reactors (MTR's) by its specific core array. The core is composed of hexagonal Be blocks with central channels (see Figure C.1). These channels form a twisted hyperboloidal bundle and hence are close together at the mid-plane but more apart at the lower and upper ends. With this array, a high fuel density is achieved in the middle part of the vessel (reactor core with a fuelled height of 760 mm) while leaving enough space at the extremities for easy access to the channel openings. In the BR-2 reactor it is possible to irradiate fissile and structural materials intended for reactors of several types. The current operating regime of the BR-2 reactor is based on five cycles of 21 days per year (i.e. approximately 105 effective full power days).

For thermal reactor experiments, experimental irradiation devices were developed and used for Light Water Reactors and High Temperature Gas Cooled Reactors. Fast reactor experience includes experimental irradiations for Gas Cooled and Liquid Metal (sodium) cooled Fast Reactors. The irradiation conditions include and often focus on off-normal and transient conditions and on safety experiments. Table C.1 summarises the essential characteristics of the 100 different irradiation positions in BR-2 and Figure C.2 indicates the variety of spectra at high neutron flux level available in the BR-2 reactor. In the same figure, an example is given of the flux spectra without (curve b) and with (curve c) cadmium screen located in the axis of a fuel element channel. By cutting off the thermal component of the neutron flux spectrum the radial fission density distributions across fuel pin bundles and inside the fuel pins themselves become much flatter, leading to temperature distributions in the pins and bundles simulating better those occurring actually in fast reactors. To obtain adequate power rating levels, the fissile fraction of the fuel generally has to be increased. It should be noted moreover that the irradiation in BR2 of fuel pins pre-irradiated in other, thermal or fast, reactors is possible and has already been performed.

The use in BR-2 of highly enriched fuel elements with high density and incorporated burnable poison allows the acceptance of a considerable amount of negative reactivity caused by strongly neutron absorbing experiments. Table C.2 gives an overview of some test rigs developed for fuel and materials irradiation in BR-2, with emphasis on those rigs that were developed for fast reactor experiments. The design and operating features of the in-pile test sections vary from simple non-instrumented baskets to sophisticated capsules or loops reproducing typical operating conditions (rod power, cladding temperature, coolant pressure and velocity, power cycling, burn-up, coolant chemistry, etc...). Rigs were also developed that allow the irradiation of damaged or vented fuel pins, equipped for the monitoring, handling and retention of fission products in out-of-pile control circuits. There is no past experience with test rigs for Pb or PbBi cooled experiments although their design and construction will be closely derived from the proven technology of sodium devices in BR2. The development of a PbBi capsule for materials properties (corrosion and LME of

structural materials under neutron irradiation) is under development at SCK•CEN (V^{th} framework programme TECLA).

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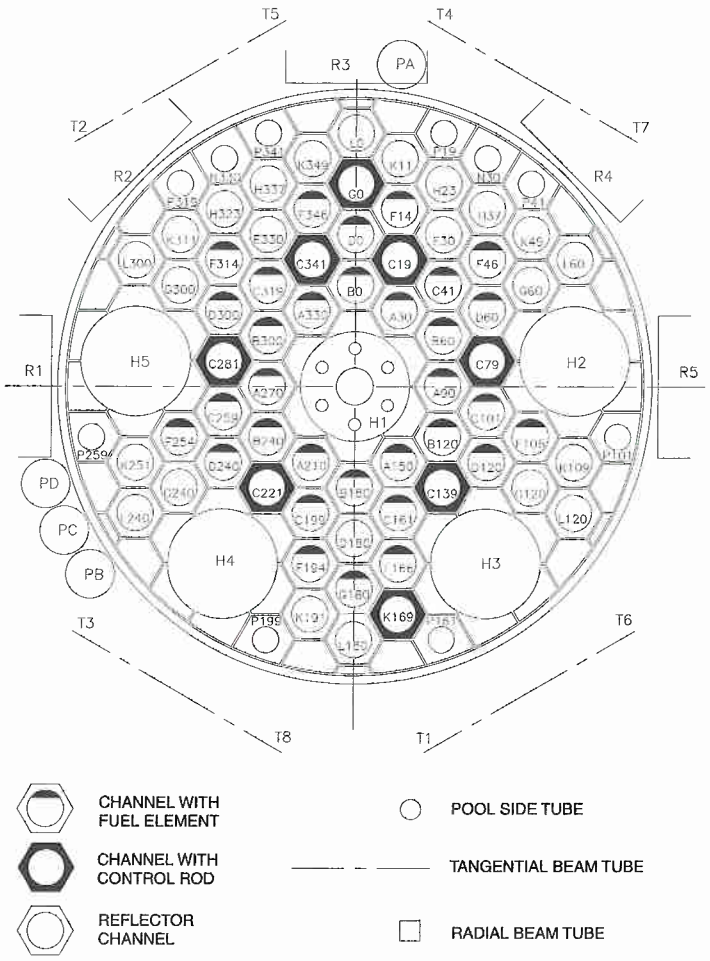

Figure C.1: Cross section of the BR 2 core at the core mid plane

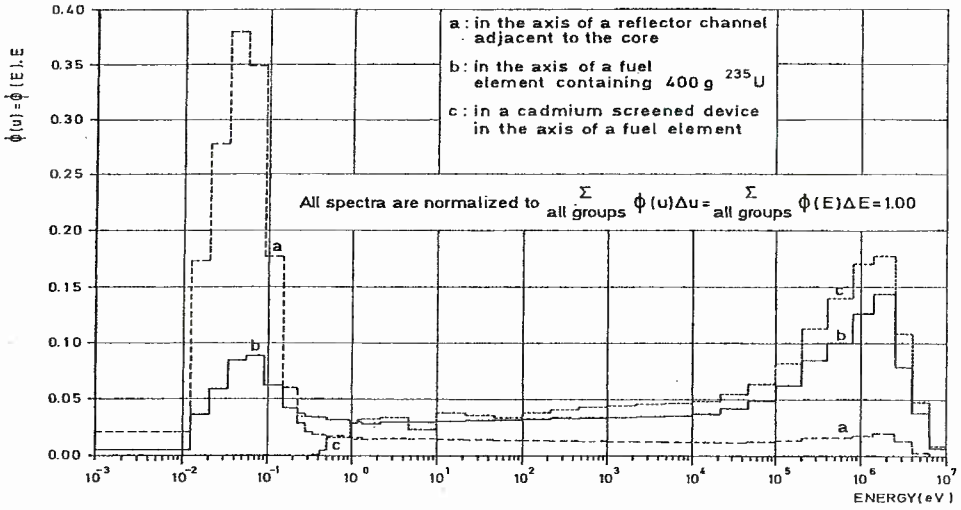

Figure C.2: Neutron spectra in different BR 2 irradiation positions

Table C.1: Characteristic data for different irradiation positions in the reactor

Position (number of positions)		Useful diameter (mm)	Max neutron flux (n/cm^2s)		Comment
			Thermal	E > 0.1 MeV	
Core region					
	Within fuel elements (30-40)	17.4 to 51.6	$2.5\text{-}4\ 10^{14}$	$5\ 10^{14}$	(b)
Reflector region					
	200 mm channel (1)	6 x 33 mm; 1x84 mm	$1.0\ 10^{15}$	$2.5\ 10^{14}$	(*)
	200 mm channels (4)	200 (3 x 84 mm)	$3.5\ 10^{14}$	$8.0\ 10^{13}$	
	84 mm channel (1)	29.5 to 80.6	$4.0\ 10^{14}$	$3\ 10^{14}$	
	84 mm channels (2)	29.5 to 80.6	$3.5\ 10^{14}$	$1.6\ 10^{14}$	(a)
	84 mm channels (20)	29.5 to 80.6	$2\text{-}3\ 10^{14}$	$<1.0\ 10^{14}$	
	50 mm channels (10)	29.5 to 46	$1.5\ 10^{14}$	3.010^{13}	

Values are given for nominal power of BR-2, which is at present 58 MWth. The reactor power varies with its core configuration and may attain a maximum of 100-120 MWth, the maximum reached in the past being 106 MWth.

(a), (b) see figure x.2 for typical neutron spectra.

(*) For dedicated experiments, this central channel is loaded with a 200 mm driver fuel element and a higher fast flux component may be obtained.

Table C.2: Non-exhaustive set of data on typical irradiation rig experience

Irradiation device	Status and Type (stagnant-circulating-with or without instrumentation)	Past or present experience and major constraints on fuel types or targets (incl. limits on MA cq. LLFP containing targets)
Gas cooling (He, Ne, CO_2)	- stagnant gas capsules and gas loops - graphite and coated particles fuel for thermal GCR's - vented fuel pins for gas cooled fast reactors - instrumentation: T, P, flow rate, fission gas release, n-flux	- over one hundred capsules and loops have been irradiated - detailed design and operational experience are available - No reactor-specific constraints on fuel targets or material targets with respect to composition, enrichment, compatibility or geometry (*). If required, double-barrier devices are used to provide adequate safety.
Na / NaK cooling	- stagnant NaK capsules and Na loops - fuel rods, fuel bundles and structural materials for LMFR and fusion reactors - instrumentation: T, P, flow rate, fission gas release, fuel failure detection, n-flux	- several hundreds of capsules and loops have been irradiated - detailed design and operational experience are available - No reactor-specific constraints on fuel targets or material targets with respect to composition, enrichment, compatibility or geometry (*). If required, double-barrier devices are used to provide adequate safety.
Pb cooling	none	none
PbBi cooling	In preparation	TECLA programme (V^{th} framework)

(*) Note: the overall geometry of the rig must of course fit in the channel dimensions in BR-2, as given in table x.1

C2. OSIRIS

OSIRIS is an open-core pool-type research reactor. Original design solutions were developed to meet six essential criteria or requirements:

- High neutron capabilities with fuel meeting international standards.
- Operational safety satisfying the most stringent requirements.
- Flexibility and ease in use and operation.
- Integrated design allowing easy transfer of OSIRIS facilities to ISIS or to hot cells.
- Complete access to core providing high irradiation adaptability and capacities.
- Provision of complementary specialized services.

The compact core (55 x 57 x 60 cm^3) is made up of 38 standard fuel elements and 6 control elements with hafnium as absorber. Very high neutron fluxes, > $3.10^{14} n.cm^{-2}.s^{-1}$, can be obtained.

- Thermal power : 70 MW
- Fuel : U_3Si_2Al enriched to 19.75%
- Moderator : H_2O
- Reflector : H_2O, beryllium
- Thermal neutron flux : $3.10^{14} n.cm^{-2}.s^{-1}$
- Fast neutron flux (> 0.1 MeV) : $4.5.10^{14} n.cm^{-2}.s^{-1}$
- Core inlet temperature : 38°C
- Core outlet temperature : 47°C
- Pool temperature : 35°C
- Primary core flow rate : 5600 $m^3.h^{-1}$
- Average heat flux : 125 $W.cm^{-2}$
- Maximum heat flux : 310 $W.cm^{-2}$
- Secondary inlet temperature : 23°C
- Secondary outlet temperature : 36°C
- Secondary core circuit flow rate : 4200 $m^3.h^{-1}$

OSIRIS is a polyvalent reactor, used for :

- Technological irradiations for nuclear industry and fundamental research (irradiation of materials for controlled fusion) requirements.
- Production of radioisotopes and doped silicon.
- Activation analysis.

The design principle of an open core pool reactor with no pressurisation vessel provides direct access to the core. The cellular core structure comprises 56 positions of which 44 are occupied by fuel elements. 16 experimental facilities can be irradiated simultaneously in the core and several in the beryllium reflector. There are 3 grids at the core periphery on 3 faces of the core structure. These make it possible to insert 27 experimental facilities at the same time in

the first periphery. The experimenters thus have a wide choice of positions depending on the desired thermal and fast neutron flux levels. These grids have been designed to accept irradiation experiments for very long (up to 2 metres) PWR fuel rods, of assemblies coming from nuclear power reactors. Displacement systems allow the adjustment of experimental device positions with respect to the core, so as to precisely adjust the irradiation conditions and to carry out power variations (cycling, ramps), representative of normal or transient conditions in nuclear power reactors. In addition, these displacement systems are designed for the insertion and removal of experimental devices with the reactor in operation.

The actual grid arrangement is given in figure C.2

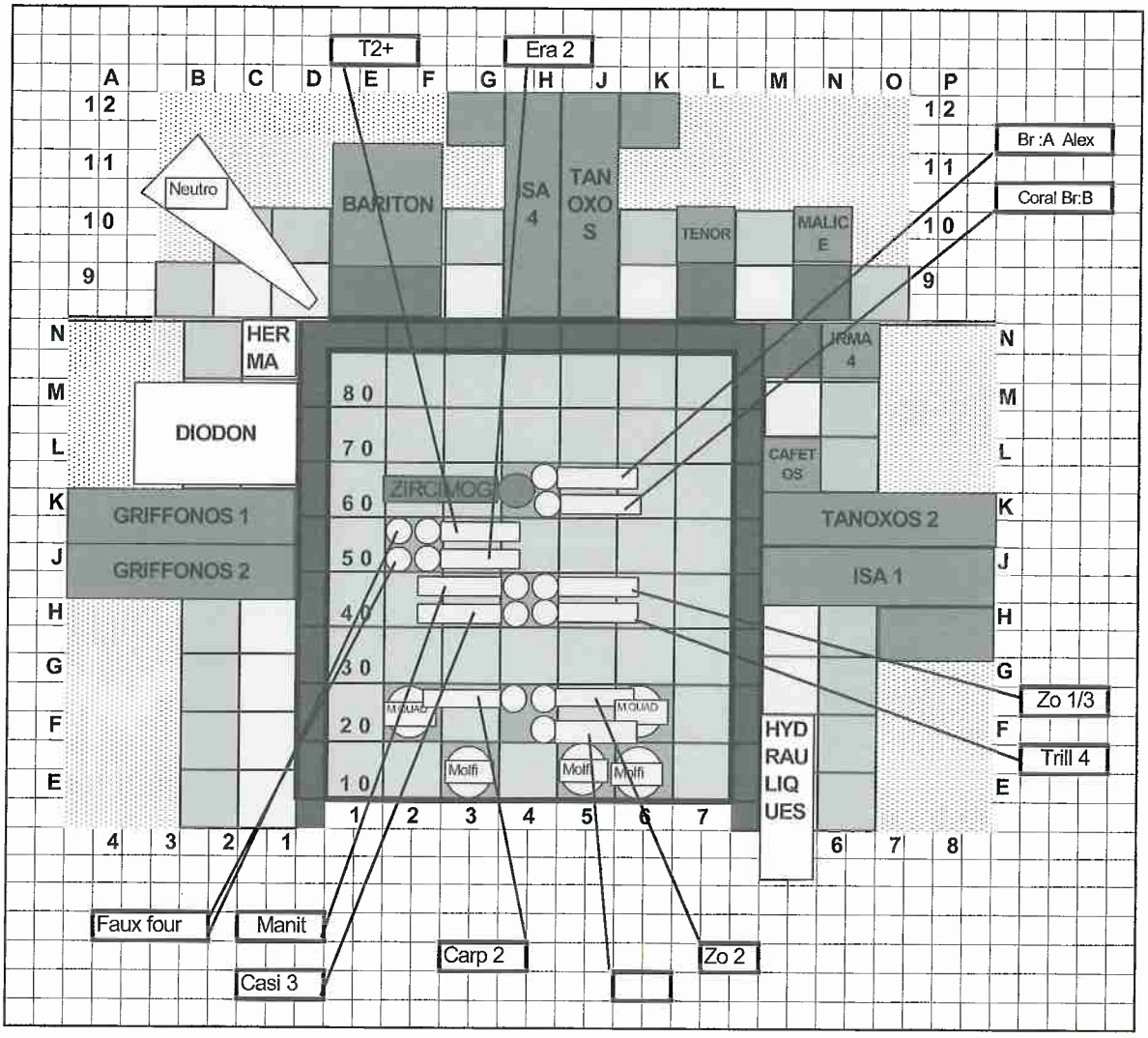

Figure C2: Core layout of the OSIRIS reactor.

Table C3: Fuel irradiation devices in OSIRIS.

	Loop for PWR OR BWR fuel		Boiling capsule for PWR or BWR fuel	Capsule for high burn-up fuel
Irradiation device	**ISABELLE 1**	**ISABELLE 4**	**GRIFFONOS**	**TANOXOS**
Existing devices	1	1	2	1
Fuel rods (fresh/preirradiated)	1	1 to 4	1	1 to 6
				(fresh)
Medium	H2O (PWR conditions)	H2O (PWR conditions)	H_2O	gas
Test channel				
Diameter	19,6 mm	26,4 x 26,4 mm	46 mm	6 x 12 mm
Length	1200 mm	2000 mm	<2000 mm	200 mm
Power maximum				
Loop	60 kW	140 kW	60 kW	65 kW
fuel rod	37 kW	90 kW	37 kW	10 kW
LHR	550 W/cm	500 W/cm	550 W/cm	550 W/cm
Precision (1□)	3%	5%		
Temperature maximum			250 °C	1000 °C
H_2O inlet	250 °C	300 °C	344 °C	80 °C
Outside fuel clad	344 °C	344 °C	± 5 °C	± 5°C
Instrumented rods	yes	no	yes	yes
				on-line monitoring of central temperature (<1000 °C)
Neutronics				
Flux level (E> 1MeV)	$3x10^{13}$ n.cm^{-2}.s^{-1}	$3x10^{13}$ n.cm^{-2}.s^{-1}	$3x10^{13}$ n.cm^{-2}.s^{-1}	$3x10^{13}$ n.cm^{-2}.s^{-1}
Power control				
Adjustment	displacement	displacement	displacement	displacement
Thermal instrumentation	yes	Yes	yes	yes
Neutronic instrumentation	yes	Yes	yes	yes
Programs				
Usual	**power ramps**	**burn-up**	separate variables studies	parametric studies for high burn-up advanced fuel
Specific	Thermal cond.	Li-Pb (fusion)	in-situ metrology	
		power cycling		
Date of commissionning	1985	1983	1998	1996

Table C4: Materials irradiation devices in OSIRIS.

	Capsule for PWR or BWR material			Capsule for LWR PV steel	Capsule for non-LWR material
Irradiation device	CHOUCA	CHOUCA NIa	CORALLINE	IRMA	COLIBRI
Existing devices	8 to 10	1	2	3	1
	passive fluence rigs	creep studies			
Medium	NaK	NaK	H_2O	Helium	Helium
			PWR conditions		
Test channel					
Diameter	21 mm	28 mm	23 mm	25 x 60 mm	34 mm
Length	400 mm	100 mm	420 mm	400 mm	400 mm
Maximum specimens					
Number	> 80	2	40 blades/4 tubes	80	> 40
Mass	200 g	20 g	200 g	8	200 g
Temperature					
Range	250 - 350 °C	300 - 350 °C	325 °C	250 - 350 °C	550 - 950 °C
Precision	± 10 °C	± 10 °C	± 8 °C	± 5 °C	± 20 °C
Electrical heaters	yes	yes	yes	yes	no (gas gap only)
Instrumentation					
Thermocouples	18	8	12	18	15
Fluence monitors	12	4	8	24	12
Neutronics					
Flux level (E> 1Mev)	$2x10^{14}$ n.cm^{-2}.s^{-1}	$2x10^{14}$ n.cm^{-2}.s^{-2}	$2x10^{14}$ n.cm^{-2}.s^{-2}	$1,4x10^{12}$ n.cm^{-2}.s^{-1}	$2x10^{14}$ n.cm^{-2}.s^{-1}
dpa/flux (barn)	1600	1600	1600	1400	1600
Programs					
Usual	Zr alloys development		clad alloys corrosion studies	PV embrittlement analysis	high temperature materials
Specific	dynamic applied stress	internal pressure induced stress		spectrum influence	fusion blanket materials
Date of commissionning	continuous	1998	1988	1992	1989

C3. R2 reactor (Studsvik)

no information received

C4. HFR Reactor (JRC)

The High Flux Reactor (HFR) Petten, managed by the Institute of Advanced Materials (IAM) of the JRC of the European Commission, is one of the most powerful multi-purpose materials testing reactors in the world.

The HFR is of the tank-in-pool type, light water cooled and moderated and operated at 45 MW. In operation since 1962, and following complete refurbishment in recent years, the HFR has a technical life beyond the year 2015.

The reactor provides a variety of irradiation facilities and possibilities: in the reactor core, in the reflector region, in the poolside. Horizontal beam tubes are available for research with neutrons. Gamma irradiation facilities are also available. Excellently equipped hot cell laboratories, on the Petten site, can provide virtually all envisaged post-irradiation examinations.

The close co-operation between the JRC and the NRG on all aspects of nuclear research and technology is essential to maintain the key position of the HFR amongst research reactors worldwide. This co-operation has led to a unique HFR structure, in which both organisations are involved with the aim to adopt a more market oriented approach and offer their long standing and recognised competence in exploiting a powerful, reliable set of nuclear facilities to world-wide interested parties.

HFR is also in the core of the Medical Valley association. This association between IAM, NRG, Mallinckrodt Medical. Urenco and hospitals leads to a medical structure unique in the world.

The operating pattern of the HFR follows a 28-days operation period, comprising 24.7 days of reactor operation, followed by a 3.3 days standard shut-down period. This shut-down period is used for installation and reloading of the irradiation rigs, maintenance to the reactor and experimental facilities, and for refuelling. This reactor is operated at 45 MW for 270 to 290 days per year. Two extended shut-down periods of approximately 4 weeks, i.e. a first period for maintenance and modifications at the beginning of the year, and a second period for maintenance, training and holidays during the summer season, interrupt the reactor operation.

The annual cycle calendar is established in close co-operation with the BR2 and Osiris reactors in order to guarantee the year around production of radioisotopes. The HFR annual cycle calendar is usually followed within a few hours.

The core lattice is a 9 x 9 arrays (729 mm x 750.4 mm) containing 33 fuel assemblies, 6 control assemblies, 19 experiment positions and 23 beryllium reflector elements. The standard core configuration is shown in Figure C3. Typical nuclear values are given for each experimental position. Some characteristic values for the HFR are presented in table C5.

Table C6 shows a number of irradiation devices that can be used for irradiation tests on ADS fuel or other types of fuel rods. This equipment can either be used for steady state or transient irradiations. All HFR fuel test facilities are included except those facilities used for MTR or HTR fuel testing. Also the irradiation facilities used in the past for in-core accident simulation have not been included.

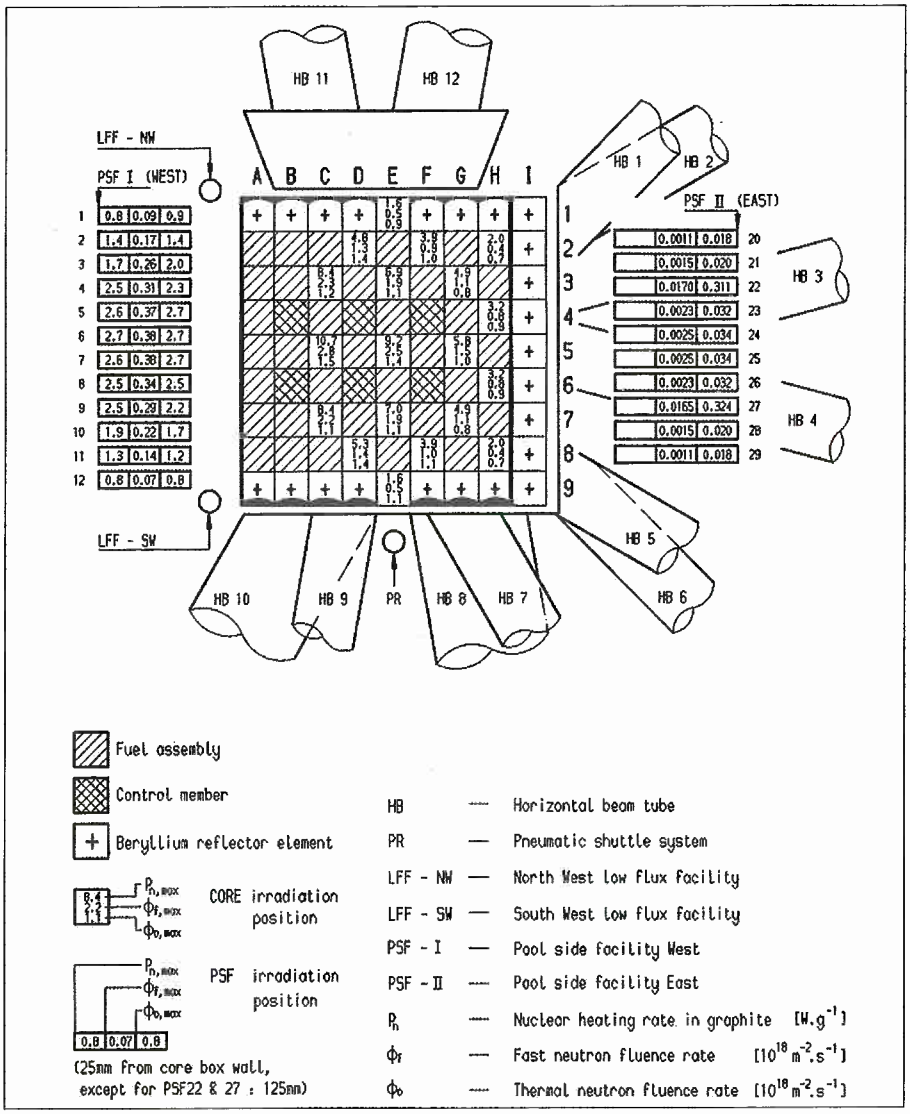

Figure C.3: Core configuration and nuclear values of the HFR

Table C.5: HFR Characteristics

Reactor thermal power	45 MW
Specific power	310 MW/m^3
Number of fuel elements	33
Number of control assemblies	6
Number of in-core irradiation positions	19
Number of pool side facility positions	22
Number of horizontal beam tubes	12
Fuel	UAl_x
Moderator	H_2O
Reflector	Beryllium
Core volume	0.15 m^3
Core inlet temperature	45°C
Core outlet temperature	55°C
Primary core flow rate	4100 $m^3.h^{-1}$
Average heat flux	100 $W.cm^{-2}$
Maximum heat flux	160 $W.cm^{-2}$
Primary system pressure at core outlet	0.34 MPa
Pressure difference across the reactor core	0.11 MPa

Table C.6: Irradiation facilities at the HFR.

Facility	TRIOX
Description	Facility used for steady state irradiation of fast reactor type fuels using a cadmium shield to harden the spectrum
Experience	FBR fuel, CAPRA fuel
Position in HFR	In-core position
Coolant	Stagnant sodium
Cladding outside temperature	400-600 °C. Axial temperature distribution can be tailored.
Fuel rod dimensions	Length < 600 mm
Fuel rod power	Typical 400 W/cm - 700 W/cm
Instrumented	Pressure transducer, thermocouple
Thermal flux	Depending on Cd configuration
Fast flux (E>0.1 MeV)	Depending on Cd configuration

Facility	KAKADU
Description	Facility used for ramp testing of fast reactor type fuels. The fuel power is varied by moving the rod with respect to the centre of the core
Experience	FBR fuel
Position in HFR	Pool Side Facility
Coolant	Stagnant sodium
Cladding outside temperature	400-600 °C. Axial temperature distribution can be tailored.
Fuel rod dimensions	Length < 2000 mm
Fuel rod power	Typical 400 W/cm - 700 W/cm
Instrumented	Pressure transducer, thermocouple
Thermal flux	$3*10^{18}$ $m^{-2}s^{-1}$ (maximum)
Fast flux (E>0.1 MeV)	$0.8*10^{18}$ $m^{-2}s^{-1}$ (maximum)

Facility	SHIFT (Sample Holder For Instrumented Fuel Tests)
Description	Facility mainly used for steady state irradiation till high burnup
Experience	Steady state irradiation of various types of fuels or transmutation targets. Examples of past irradiations are: AmO_x in $MgAl_2O_4$ UN (U,Pu)N UO_2 in inert matrics (a.o $MgAl_2O_4$ and MgO) $(U,Pu)O_2$ $(Zr,Y,Pu,Er)O_2$ $(Zr,Y,Pu,Er)O_2$ in $MgAl_2O_4$
Position in HFR	In-core position
Coolant	Stagnant sodium or aluminium drum
Cladding outside temperature	300 - 600 °C
Fuel rod dimensions	Length < 600 mm
Fuel rod power	Maximum 1000 W/cm
Instrumented	Pressure transducer, thermocouple
Thermal flux	$1.5*10^{18}$ $m^{-2}s^{-1}$ (maximum)
Fast flux (E>0.1 MeV)	$4.6*10^{18}$ $m^{-2}s^{-1}$ (maximum)

Facility	BWFC (Boiling water fuel capsule)
Description	Facility used for ramp testing or short-term irradiation under PWR/BWR conditions. Fuel power is changed by varying position of fuel rod with respect to reactor core.
Experience	Mainly ramp testing commercial LWR fuel rods
Position in HFR	Pool Side Facility
Coolant	Stagnant water, with either PWR or BWR typical temperature and pressure
Cladding outside temperature	LWR typical
Fuel rod dimensions	Outer diameter < 22 mm Typical 400 mm
Fuel rod power	Typical 400 W/cm - 700 W/cm
Instrumented	Pressure transducer, thermocouple
Thermal flux	$3*10^{18}$ $m^{-2}s^{-1}$ (maximum)
Fast flux (E>0.1 MeV)	$0.8*10^{18}$ $m^{-2}s^{-1}$ (maximum)

Facility	MOKA
Description	Facility used for base irradiation in an in-core position under PWR/BWR conditions. After base irradiation the facility can be moved from an in-core position to the Pool Side Facility in which a ramp test can be performed, by varying the position of fuel rod with respect to reactor core, similar to the above-mentioned BWFC.
Experience	LWR fuel rods
Position in HFR	Base-irradiation in-core, ramp test Pool Side Facility
Coolant	Stagnant water, with either PWR or BWR typical temperature and pressure
Cladding outside temperature	LWR typical
Fuel rod dimensions	Outer diameter < 22 mm Typical 400 mm
Fuel rod power	Typical 400 W/cm - 700 W/cm
Instrumented	Pressure transducer, thermocouple
Thermal flux	$3*10^{18}$ $m^{-2}s^{-1}$ (maximum PSF) $1.5*10^{18}$ $m^{-2}s^{-1}$ (maximum in-core)
Fast flux (E>0.1 MeV)	$0.8*10^{18}$ $m^{-2}s^{-1}$ (maximum PSF) $4.6*10^{18}$ $m^{-2}s^{-1}$ (maximum in-core)

Facility	POTRA (Power TRAnsient)
Description	Facility used for ramp testing of fast reactor type fuels. The power is varied by changing the gas pressure in a BF_3 filled gas compartment surrounding the fuel pin.
Experience	FBR fuel
Position in HFR	In-core
Coolant	Stagnant sodium
Cladding outside temperature	400-600 °C. Axial temperature distribution can be tailored.
Fuel rod dimensions	Length < 2000 mm
Fuel rod power	Typical 400 W/cm - 700 W/cm
Instrumented	Pressure transducer, thermocouple
Thermal flux	$1.5*10^{18}$ $m^{-2}s^{-1}$ (maximum)
Fast flux (E>0.1 MeV)	$4.6*10^{18}$ $m^{-2}s^{-1}$ (maximum)

C5. THE HALDEN HBWR REACTOR

The HBWR is a natural circulation boiling heavy water reactor situated within a rock hillside in Halden, Norway. The maximum power is 25 MW (th), and the coolant/moderator temperature is 240°C, corresponding to an operating pressure of 33.3 bar.

The flat reactor lid has more than 300 penetrations for fuel assemblies, control stations and experimental equipment. The wide core gives good room for instruments and experimental equipment. Light water, high pressure loops provide facilities for testing under prototypic BWR and PWR conditions.

The core consists of about 110 fuel assemblies, including up to 40 instrumented test fuel assemblies, in an open hexagonal lattice with a lattice pitch of 130 mm. 30 lattice positions are occupied by control stations. The maximum height available for insertion of experimental fuels and materials is 1710 mm. Selected core data are given in Table C.7, and Fig. C.4 shows a core map which is typical of a core for the HBWR experimental programme.

Table C.7. Selected Core Data for the Halden Reactor

Number of Fuel Assembly	110
Number of Control Stations	30
Core height	1710 mm
Height of driver fuel	800 mm
Configuration	open hexagonal lattice
Lattice Pitch	130 mm
Reflector Top: thickness	300 mm
Reflector Bottom: thickness	380 mm

There are normally 2 main shutdowns per year, dictated primarily by the experimental programmes. Two operation periods of 95 - 100 full power days are aimed at per year, resulting in an availability of about 55%. The basis for the experimental programme performed in the HBWR is a range of experimental circuits, test rigs, and in-core instruments developed and designed by the Halden Reactor Project.

Fuel and material testing under simulated water reactor conditions is performed in in-core pressure flasks connected to light water circulation systems. These systems, which are completely separated from the reactor cooling systems, are designed for operation at pressures and temperatures at 165 bar and 340°C, respectively. In-core pressure flask can be surrounded by high enriched fuel rods in order to obtain neutron flux conditions typical of other types of reactors.

An Instrumented Fuel Assembly (IFA) represents a test rig in which a set of fuel rods or material specimens can be irradiated. In most cases the test rigs are designed such that fuel rods and specimens can be exchanged, thus facilitating multipurpose use and economy.

Typical flux spectra for channels with and without fuel (but filled with steel specimens) are shown in Fig. C.5. Other spectra can be obtained in reflector channels or by introducing absorbers. Maximum values for fast and thermal flux in typical channels are given in Table C.8 together with available space in these channels and their number.

Several experimental circuits are permanently installed to support testing in special rigs:

- He-3 flux control system
- diameter gauge drive systems
- fuel rod drive system
- fuel rod gas flow system
- gas supply system
- ultra high gas pressurising system

The range of experiments in recent years and currently includes testing of re-fabricated commercial high burnup fuel, cladding alloys, core structural materials as well as control materials. Although design and feasibility studies related to testing of fast breeder fuel have been performed in the past, no irradiation experience exists involving liquid metals. However, the possibility of ADS related irradiation of materials in PbBi cooling is being assessed for a participant of the Halden Project. An overview of experience with fuels for different reactor types is given in Table B9.

Table C.8: Characteristic data for different irradiation positions in the reactor

Position/assembly type	Useful Ø (mm)	Max neutron flux (n/cm^2s) thermal	E>0.1 MeV	Number of position
Core region				
Unfuelled channel	71	1.2×10^{14}	1×10^{13}	40[a)]
Within driver fuel	40	5×10^{13}	5×10^{13}	70
Booster fuel with pressure flask	40	4×10^{13}	1.6×10^{14}	15[b)]
Instrumented fuel assembly	71	$3\text{-}10 \times 10^{13}$	$2\text{-}10 \times 10^{13}$	40[c)]
Reflector channel	71	$<1 \times 10^{14}$	$0.01 - 1 \times 10^{13}$	many

a) any position within the core

b) any position within the core, total given by capacity of installed loops.

c) any position within the core, sum of (a) + (b) + (c) in use for experiments about 35 - 40

Table C.9: Irradiation rig experience

Irradiation device [1)]	Status and Type	Past and present experience
Instrumented assemblies in loops	several loops in operation, numerous applications	PWR, BWR, ATR, CANDU conditions
Gas cooled instrumented assembly	loop in operation	Testing of gas-cooled reactor fuels and component materials
Irradiation capsules	- sealed capsules - capsules allowing gas exchange - instrumented (temperature, pressure, expansion, failure detection, sampling lines)	Dry irradiation of materials at typical temperatures; Irradiation of fuels of gas cooled reactors at elevated temperatures
PbBi cooling	in preparation	none

Note 1) All experimental set-ups are contained in irradiation rigs with dimensional constraints as indicated in Table B6. These rigs contain the nuclear instrumentation (flux monitors) and coolant related instrumentation (temperature, flow).

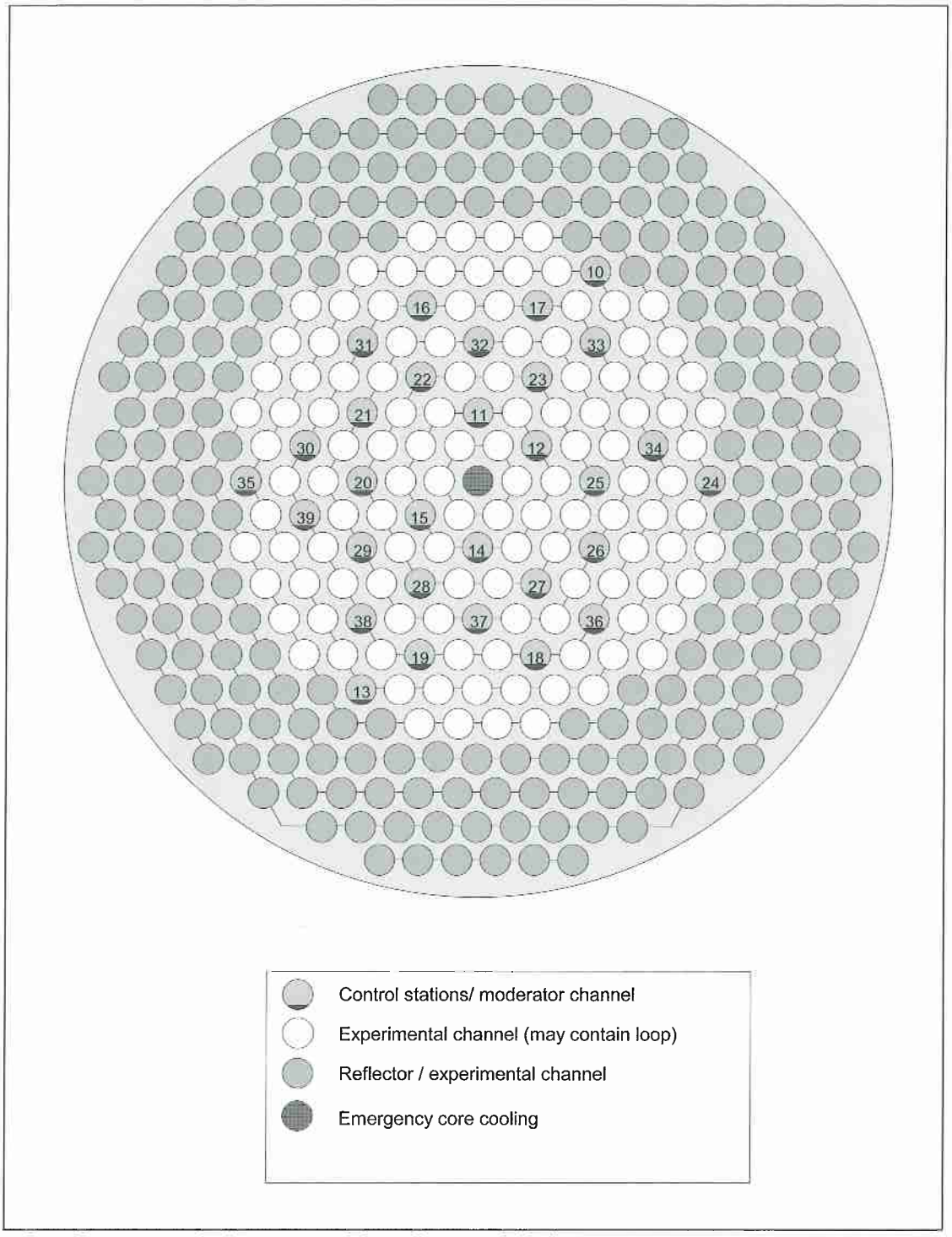

Fig. C.4: Cross-section of the HBWR core.

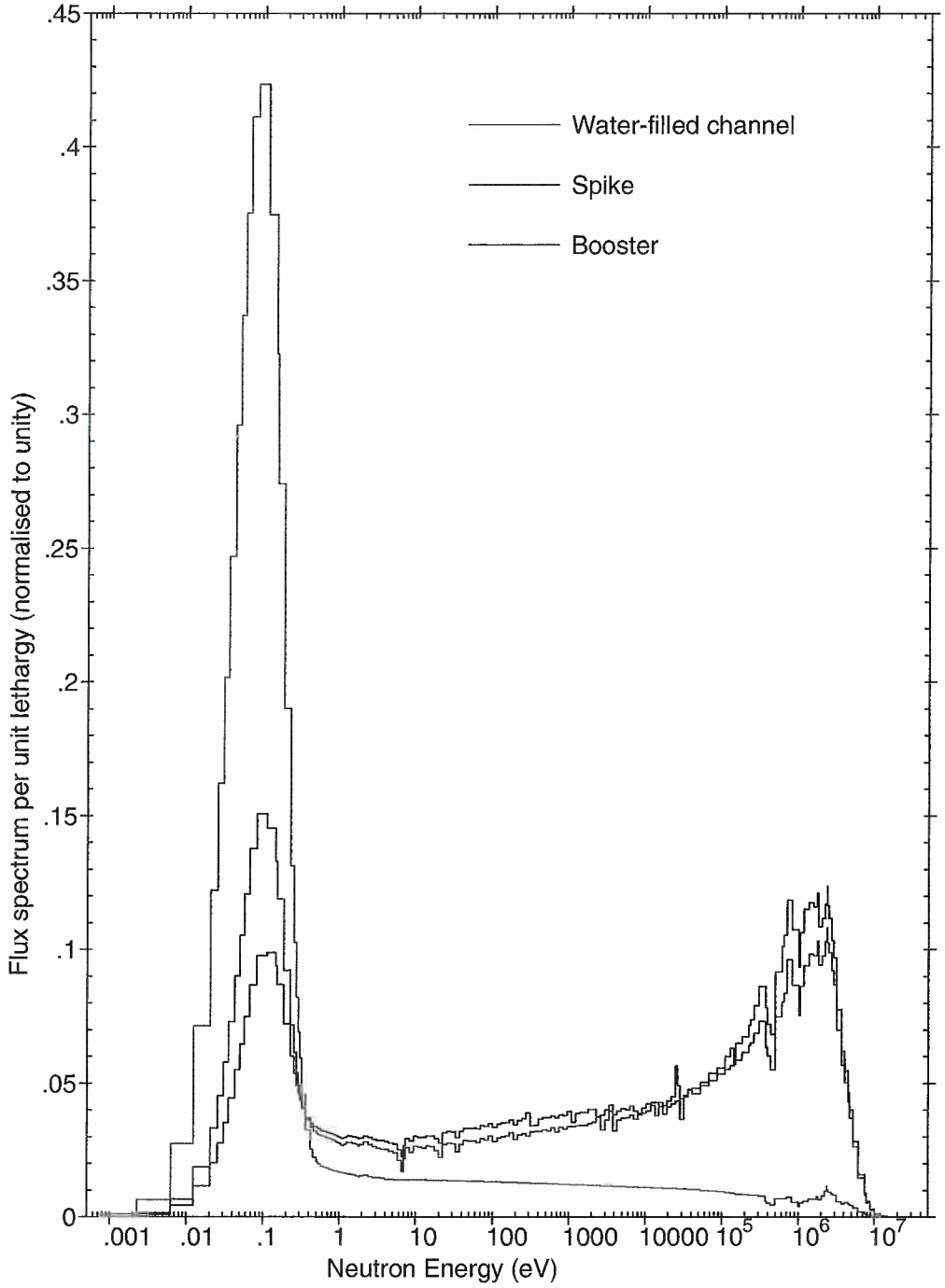

Fig. C.5: Example of flux spectra in different channels of the HBWR.

Appendix D: European Hot-cell facilities

CEA	
Addresses	LECA/STAR CEA/CADARACHE F-13108 St-Paul-Lez-Durance cedex LAMA (until 2007) (*) CEA/GRENOBLE 17, rue des Martyrs F-38054 Grenoble cedex 9 LECI (*) CEA/SACLAY F-91191 Gif-sur Yvette cedex
Responsible	Mrs. M. Tourasse (LECA/STAR and LAMA) Mr. Ph. Billot (LECI)
Main purpose of facility	PIE of fuel rods, targets (LECA/STAR and LAMA) and structural materials (LECI)
Number and type of cells in LECA/STAR	10 concrete / 5 lead shielded [(a)] in LECA – 3 concrete in STAR [(b)]
Number and type of cells in LAMA	11 concrete / 7 hot laboratories equipped with lead shielded cells and/or glove boxes
PIE techniques – NDT	Visual - dimensional control (length, inner and outer diameter) – EC testing – X-radiography – □ spectrometry
PIE techniques – DT	Optical microscopy – SEM – EPMA - SIMS– α/β autoradiography - XRD - TEM(°) – density – open porosity – radiochemical analysis (mass spectr., gas chrom.) - rod puncture - clad hydrogen analysis - clad burst testing - clad corrosion - fuel creep testing - fuel heat treatment
Refabrication	Rod segments refabrication including He-filling, pressurization and leak and XR testing
Acceptance of failed rods	Yes
γ -activity limits	40 / 400 / 4000 / 40000 TBq in concrete cells in LECA (0.8 / 40 TBq in lead shielded cells)
Cell characteristics (glove box in shielded cell)	In 1 cell in LECA (refabrication - 2 others are going to be equipped in 2003) – The 3 of STAR
Maximum length of rod which can be handled	1400 MW PWR fuel rod type (little more than 4 m)
Acceptance of Minor Actinides (Np, Am, Cm)	In 2003 (when equipped) – Minor actinides as trace elements accepted presently
Experience with Minor Actinide research	No, except 1 irradiated experimental FBR bundle (SUPERFACT in 1990 – open litt. available)
Acceptance limits of Pu	350 g in U+Pu (with no H moderator limit), 2.5 kg (with 5 l max of mod.), 10 kg (with no mod.)

(*) Details about these laboratories are witheld from the present summary review in view of their restrictions (LAMA will be out of operation in 2007, LECI accepts only structural materials)

(°) Restrictions on activity and contamination level apply

(a) to be dismantled in 2005

(b) 1 devoted to the spent Magnox fuel conditioning

CIEMAT	
Address	Avda. Complutense, 22 28040 Madrid Spain
Responsible	José Luis Díaz Díaz (jl.didiaz@ciemat.es)
Main purpose of facility	PIE of structural materials **(no fuel) (*)**
Number and type of cells	
PIE techniques - NDT	
PIE techniques - DT	
Refabrication	
Acceptance of failed rods	
γ -activity limits	
Cell characteristics	
Maximum length of rod which can be handled	
Acceptance of Minor Actinides (Np, Am, Cm)	
Experience with Minor Actinide research	
Acceptance limits of Pu	

(*) Further details about this laboratory are withheld from the present summary review in view of its restrictions (CIEMAT accepts only structural materials)

IFE	
Facility location	Institutt for energiteknikk, ife, P.O.B. 40, N-2007 Kjeller, Norway
Responsible	Knut.Lunde (knut.lunde@ife.no)
Contact person	Dr.-Ing. B.C.Oberländer (barbara.oberlander@ife.no)
Main purpose of facility	PIE and refabrication of fuel rods and structural components
Number and type of cells	3 concrete cells, 3 lead cells
PIE techniques – NDT	Visual – dimensional control (length and diameter) – EC testing – □ spectrometry- neutronradiography
PIE techniques – DT	Optical microscopy – SEM(°)- α/β autoradiography - O/M-ratio – density - radiochemical analysis of irradiated fuel - rod puncture - clad hydrogen analysis – clad tensile testing - clad burst testing - micro hardness
Refabrication and instrumentation	Refabrication: since 1991 Instrumentation in hot cell : since 1991
Acceptance of failed rods	failed rods with auxiliary cladding
γ -activity limits	concrete cells: 4 PBq
Cell characteristics	3 concrete cells with no inner glove boxes, 3 lead cells with inner glove boxes-
Maximum length of rod which can be handled	2.5 m (exception: 4m)
Acceptance of Minor Actinides (Np, Am, Cm)	as present in irradiated fuel
Experience with Minor Actinide research	Cm-no; Am-chemical separation+α; Np-no
Acceptance of fissile material (U, Pu)	80 % of lowest critical mass *, max fissile content: 20%**
Experience with Pu research	fuel pellets

(°) Restrictions on activity and contamination level apply

(*)with full moderation and reflection, (**) U235 and/or fissile Pu

ITU – HOT CELL LABORATORY	
Address Facility location	European Commission Joint Research Centre Institute of Transuranium Elements P.O. Box 2340, 76125 Karlsruhe GERMANY Forschungszentrum Karlsruhe site
Responsible	Dr. J-P. Glatz (glatz@itu.fzk.de)
Main purpose of facility	PIE of fuel rods, targets and structural materials
Number and type of cells	19 concrete / 5 lead shielded
PIE techniques – NDT	Visual – dimensional control (length and diameter) – EC testing – X-radiography – □ spectrometry
PIE techniques – DT	Optical microscopy – SEM – EPMA – αβγ autoradiography – XRD μ-XRD – TEM (*) – density – open porosity – radiochemical analysis of irradiated fuel – rod puncture clad hydrogen analysis – clad creep testing – clad burst testing
Refabrication	Rod segments refabrication including He-filling, pressurization and leak testing
Acceptance of failed rods	No
γ-activity limits	37 PBq
Cell characteristics	11 cells are equipped with stainless steel containers, 2 with glove boxes leak rate <2-4.10^{-3} (h^{-1})
Maximum length of rod which can be handled	4.5 m
Acceptance of Minor Actinides (Np, Am, Cm)	Regulations admit minor actinides as present in irradiated materials
Acceptance of fissile material	Max.: 3500g Pu., 6000g U-235

(*) Restrictions on activity and contamination level apply

NRG•HCL	
Facility location	NRG•HCL Westerduinweg 3 1755 LE Petten The Netherlands
Responsible	Ir. A.M Versteegh (versteegh@nrg-nl.com)
Contact person	Ir. K.A. Duijves (duijves@nrg-nl.com)
Main purpose of facility	PIE of fuel and construction materials; isotope production
Number and type of cells	5 concrete /28 lead shielded
PIE techniques - NDT	Visual - dimensional control (length and diameter) - EC testing - X-radiography - γ-spectometry - γ-tomography -micro-gammascanning
PIE techniques - DT	Optical microscopy - SEM - EPMA- α/β autoradiography - TEM (*) - density - radiochemical analysis of irradiated fuel - rod puncture - tensile testing - burst testing - creep testing - charpy testing
Acceptance of failed rods	Yes
γ-activity limits	3.7 P Bq (10^5 Ci)
Cell characteristics	13 hot cells are equipped with inner glove boxes
Maximum length of rod which can be handled	4.5 m
Acceptance of Minor Actinides (Np, Am, Cm)	Only unirradiated
Experience with Minor Actinide research	Dedicated actinides laboratory is present
Acceptance of Pu	Max 17 kg (laboratory)
Experience with Pu research	LWR and MOX: extensive chemical and physical PIE research, preparation of inert matrix fuels

(*) Restrictions on activity and contamination level apply

PSI (Paul Scherrer Institute)	
Facility Location	Paul Scherrer Institute Laboratory for Materials Behaviour CH-5232 Villigen PSI Switzerland
Responsible	Dr. Gerhard Bart (gerhard.bart@psi.ch)
Contact person	Dr. Friedrich Groeschel (friedrich.groeschel@psi.ch)
Main purpose of facility	PIE of fuel rods, core components, accelerator targets, preparation of actinide ceramics
Number and type of cells	6 concrete (1 with inner glove box), 4 lead/steel shielded with inner glove box, glove box lines
Max length of rods	4.5 m
PIE techniches - NDT	optical microscopy, dimensional control (length and diameter), EC testing, cold gap measurement, gamma spectrometry, neutron radiography
PIE techniques - DT	Optical microscopy, microhardness, SEM(*), EPMA, SIMS, TEM, XRD(*), neutron diffraction, density, open porosity, ICP-MS, rod puncture, hydrogen hot gas extraction, clad tensile testing, clad burst testing, tensile testing of small specimens, □□□-autoradiography
Refabrication and instrumentation	no
Acceptance of failed fuel rods	yes
γ activity limits	NDT of fuel rods: ≤4E15 Bq fission products, NDT and DT in concrete cells: ≤1E14 Bq activation products Instrumental analysis in lead shielded cells (SIMS, EPMA): ≤4E9 Bq α,γ-activity
Acceptance of Minor Actinides (Np, Am, Cm)	Unirradiated samples in glove boxes: Np-237: ≤5E8 Bq, Am-241: ≤1E11 Bq Irradiated samples: accepted with special safety case
Experience with Minor Actinides	Preparation of $(U,Pu,Np)O_2$ ceramics completed
Acceptance of Pu	Unirradiated material: ≤230 g Irradiated fuel: ≤20 LWR-MOX fuel pins
Experience with Pu research	LWR - and FBR fuel preparation (in oxide, nitride and carbide-form) on a lab. scale Preparation of (Pu,Zr)O2 and (Pu,Zr)N

(*) only limited activity

SCK·CEN	
Facility location	SCK·CEN LHMA Boeretang, 200 B-2400 Mol (Belgium)
Responsible	Dr. E. van Walle (evwalle@sckcen.be)
Contact person	Dr. L. Sannen (lsannen@sckcen.be)
Main purpose of facility	PIE of fuel rods and core components
Number and type of cells	8 concrete / 25 lead shielded
PIE techniques - NDT	Visual - dimensional control (length and diameter) - EC testing - X-radiography - □ spectrometry
PIE techniques - DT	Optical microscopy - SEM - EPMA - a/b autoradiography - XRD(*) - TEM (*°) - density - open porosity - radiochemical analysis of irradiated fuel - rod puncture clad hydrogen analysis - clad tensile testing (°) - clad burst testing (°)
Refabrication and instrumentation	In preparation: refabrication: due date end 2000 Instrumentation in hot cell : due date end 2002
Acceptance of failed rods	yes
γ -activity limits	2000TBq / 37TBq / 8 TBq
Cell characteristics	The hot cells have an inner glove box; leak rate $<10^{-2}$ (h^{-1}) (reference: ISO 10648-2: level 3)
Maximum length of rod which can be handled	4.5 m
Acceptance of Minor Actinides (Np, Am, Cm)	Current exploitation regulations admit minor actinides as present in irradiated fuel
Experience with Minor Actinide research	Chemical analysis (TIMS, □□ and □□ spectrometry and ICP-MS) on MA isotope production in nuclear fuel
Acceptance of Pu	Max 4kg (laboratory)
Experience with Pu research	LWR and FR MOX Extensive chemical and physical PIE research (all physical forms: solid, in solution, powder)

(*) Restrictions on activity and contamination level apply

(°) Currently (year 2000) in refurbishment

Studsvik	to be completed
Facility Location	Stusvik AB SE-61182 Nyköping Sweden
Responsible	P. Larsson (+46 155 22 16 83)
Contact person	Mr P. Askeljung (tel. +46 155 22 17 46)
Main purpose of facility	PIE of power reactor fuel and core materials
Number and type of cells	7 concrete cells (for γ-activity).
Max length of rods	<6 m
PIE techniches - NDT	Vis. insp., gamma-scan, neutron radiogr., EC, dimensional,
PIE techniques - DT	Optical and SEM, EPMA, density, chemical analysis, mechanical tests
Refabrication and instrumentation	Test rod refabrication
Acceptance of failed fuel rods	Yes
γ-activity limits	500 TBq Co-60 or equivalent
Acceptance of Minor Actinides (Np, Am, Cm)	Limited amounts, only in intimate mixture with gamma-emitters (e.g. irradiated)
Experience with Minor Actinides	Chemical analysis in low concentrations
Acceptance of Pu	Only in intimate mixture with gamma-emitters (e.g. irradiated MOX)
Experience with Pu research	Yes but not in this facility

UKAEA - Windscale	
Facility Location	Buildings B13 and B14 Windscale Seascale Cumbria CA20 1PF UK.
Responsible	Mr R Currie (ron.currie@aeat.co.uk)
Contact person	Dr R Gomme (robin.gomme@aeat.co.uk)
Main purpose of facility	PIE of fuel and reactor components Mechanical testing of components Handling/re-packaging of active materials
Number and type of cells	13 concrete shielded cells (B13) plus various smaller lead cells (B14)
Max length of rods	Typical cell dimensions: 10.5m (long) x 2.6m (wide) x 3.5m (high)
PIE techniches - NDT	Visual exam, profilometry, eddy current testing, gamma scanning,
PIE techniques - DT	Optical microscopy, SEM, EPMA,XRD, TEM, rod puncture, density, thermal diffusivity (laser flash), specific heat (differential scanning calorimetry), cladding hydrogen analysis (HVE), fatigue testing, corrosion/stress corrosion testing, impact tests, tube burst tests, crack growth tests
Refabrication and instrumentation	N/A
Acceptance of failed fuel rods	Yes
Gamma activity limits	Typical cell activity inventory $1x10^{18}$Bq
Acceptance of Minor Actinides (Np, Am, Cm)	Yes
Experience with Minor Actinides	
Acceptance of Pu	Can accept full length MOX rods
Experience with Pu research	Comprehensive MOX PIE programmes undertaken for UK and overseas customers

LIST OF ABBEVIATIONS

ADS	Accelerator-driven system
AEAT	AEA-Technology (UK)
ALI	Annual Limit on Intake
An	Actinide
ANL	Argonne National Laboratory
ATW	Accelerator-driven Transmutation of Waste
BNFL	British Nuclear Fuel plc (UK)
CAPRA	Consommation Acrue de Plutonium dans réacteurs Rapide
CDA	Core Disruptive Accident
CEA	Commissariat a l'Enegir Atomique (F)
CERN	Conseil Européen pour la Recherche Nucleaire (CH)
CIEMAT	Centro de Investigaciones Energeticas MediaAmbientales y Technologicas (E)
CRIEPI	Central Research Institute of the Electric Power Industry (J)
DOVITA	Dry reprocessing, oxide fuels, vibropac, integral, transmutation of actinides
EBR	Experimental Breeder Reactor (Argonne, USA)
EFTTRA	Experimental Feasibility of targets for Transmutation
ENEA	Ente per la Nuove Tecnologie l'Energia e l'Ambiente (I)
EU	European Union
EC	European Commission
FNR	Fast Neutron Reactor
FP	Fission Products
HFR	High Flux Reactor (Petten, NL)
HLW	High Level Waste
HLLW	High Level Liquid Waste
HM	Heavy Metal
HTGR	High Temperature Gas-cooled Reactor
IFR	Integral Fast Reactor
ICRP	International Committee on Radiation Protection
ITU	Institute for Transuranium Elements (EU)
IMF	Inert matrix Fuel
IMMOX	Inert Matrix Mixed Oxide
IVTAN	Institute for High Temperature Physics (Russia)
JAERI	Japanese Atomic Energy Research Institute (J)
JNC	Japanese Nuclear Fuel Cycle Development
JRC	Joint Research Centre (EU)
KNK-II	Kompakte Natriumgekühlte Kernreactoranlage Karlsruhe
LLFP	Long-lived Fission Products

LWR	Light Water Reactor
MFCI	Mechanical Fuel Cladding Interaction
MA	Minor Actinides
MOX	Mixed Oxide
NEA	Nuclear Energy Agency of OECD
NRG	Nuclear Research and Consultancy Group (NL)
OECD	Organisation for Economic Co-operation and Development
PIE	Post Irradiation Examinations
PSI	Paul Scherrer Institute (CH)
PUREX	Plutonium and Uranium Recovery by Extraction
PWR	Pressurised Water Reactor
RIAR	Research Institute of Atomic Reactors (Russia)
SCK·CEN	Studiecentrum Kernenergie (B)
SPIN	SeParation-INcineration
TIG	Tungsten Inert Gas
TRABANT	Transmutation and burning of actinides
TRU	Transuranium Elements
YSZ	Yttria-stabilised Zirconia

European Commission

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For the fuel cycle of accelerator-driven systems so-called advanced fuels are envisaged. These advanced fuels are characterised by a high content of plutonium and minor actinides, are preferably uranium-free and will be irradiated to relatively high burnup to achieve a high extent of transmutation. In order to select the appropriate fuel material (metal, oxide, nitride, etc.) and the fuel form (pellet, particle) the effects of the choice of the reactor coolant (gas or liquid metal) and, in case of a multiple recycle strategy, the possibility of reprocessing must be taken into account. In the present report the aspects of fuel fabrication, fuel behaviour and fuel processing are discussed. Also a review of made of the required infrastructure for development of such fuels.

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