DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS : RECHERCHES ACTUELLES

FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

Fuels and targets for transmutation

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Abstract   The major technological aspects of fuels and targets for the transmutation of minor actinides are discussed. The changes in the fuel properties, the effects on the fabrication processes and the irradiation behaviour caused by recycling of minor actinides (compared to conventional uranium-based fuel) are outlined. The most promising developments are identified. ***To cite this article: R.J.M. Konings, D. Haas, C. R. Physique 3 (2002) 1013–1022.***
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Combustibles et cibles pour la transmutation

Résumé   Les principaux aspects technologiques relatifs aux combustibles et cibles pour la transmutation des actinides mineurs sont discutés. Les modifications des propriétés du combustibles, ainsi que les conséquences du recyclage des actinides mineurs sur les procédés de fabrication et sur le comportement sous irradiation (par comparaison avec les combustibles conventionnels basés sur l'uranium) sont décrites. Les développements les plus prometteurs sont identifiés. ***Pour citer cet article : R.J.M. Konings, D. Haas, C. R. Physique 3 (2002) 1013–1022.***
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1. Introduction

Transmutation of long-lived radionuclides is a potential technology for the treatment of spent fuel from the nuclear fuel cycle. In the transmutation process the long-lived radionuclides are transformed by nuclear reactions into short-lived or stable nuclides. This can be achieved by a neutron capture reaction or by neutron capture followed by fission, normally in nuclear reactors. The former process is generally used for the transmutation of fission products, the latter for (transuranium) actinides.

For the transmutation process the long-lived must be separated from the short-lived radionuclides and since most radionuclides have different optimum transmutation conditions, the separation of the long-lived radionuclides from each other may be required also. Transmutation is therefore always coupled
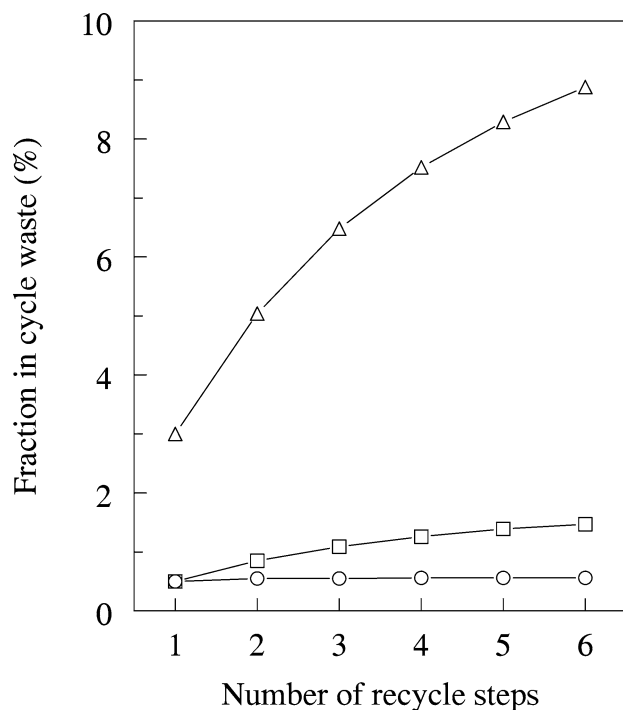

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Figure 1. The fraction of actinides in the cycle waste as a function of the number of recycle steps. ○, partitioning efficiency (ε_p) is 99.5% (including fabrication wastes) and transmutation efficiency (ε_t) is 90%; □, $\varepsilon_p = 99.5\%$, $\varepsilon_t = 30\%$; △, $\varepsilon_p = 97\%$, $\varepsilon_t = 30\%$.

to the reprocessing of spent fuel, but one normally speaks of partitioning to indicate the separation of radionuclides other than Pu and U. The overall efficiency of any transmutation strategy is thus the product of the destruction rate in the reactor and the efficiency of the partitioning process (Fig. 1). Therefore extensive research on the reactor design and on the development and optimisation of new separation processes is in progress [1].

Also the design and fabrication of fuels or targets from the separated radionuclides are key elements in the transmutation strategy. This needs special attention, because the fabrication and irradiation behaviour are significantly different from UO_2 fuel, as we will discuss in the present paper. We will focus on fuels and targets of transuranium elements, as these elements pose the most serious problems due to their high radiotoxicity. In this context, targets are materials whose fissile/fertile composition does not contribute significantly to the criticality of the reactor core.

Numerous fuel and target concepts have been proposed for the transmutation of transuranium actinides, including various older concepts that are re-visited. The variety of proposals reflects the technical complexity due to the many different requirements coming from strategy studies. The fuel choice often reflects specific fuel cycle aspects such as fabrication, irradiation behaviour or reprocessing, which all have their own technological difficulties. On top of this comes, of course, the choice of reactor, which can have a fast or thermal neutron spectrum, can be gas-cooled or liquid–metal cooled, etc. Irradiation in fast reactors is generally considered to give the best transmutation efficiency (eventually also in thermalised zones), not only because of the fast spectrum but also because of the neutron economy. The latter is even better in accelerator-driven systems.

In the present paper we will briefly discuss the most important solid fuel and target forms, giving special attention to the following specific problems associated with transmutation fuels and targets:

- the changes in the chemical and physical properties (thermal conductivity, chemical stability) going from uranium compounds to americium and curium compounds;

- the high radiotoxicity of especially americium and curium and the concomitant difficulties for fabrication and processing;
- the relatively large helium production in the transmutation chains of Am and Cm, compared to uranium or U, Pu fuels;
- the difficulties to reprocess certain transmutation fuels and targets.

2. Physico-chemical properties of transuranium fuels and targets

2.1. Homogeneous fuels and targets

The properties of the actinide metals change drastically along the actinide series, which is partly related to the different metal valence states: tetravalent (Th–Pu), trivalent (Ac, Am–Cf, Lr) and divalent (Es–No). For example, the melting point of the actinide metals decreases about 500 K going from U to Pu (Fig. 2). Therefore, the melting point of a metal fuel containing large amounts of transuranium actinides is significantly lower than that of pure uranium metal fuel. Addition of a significant quantity of a non-fissile metal with a high melting point is thus required. Zirconium (melting point 2128 ± 5 K) is the most likely candidate, as it is already added to uranium fuel to suppress anisotropic swelling at low burn-up [2]. However, there is considerable uncertainty about the (mutual) solubility of neptunium and zirconium for which the existing experimental studies are in contradiction. The vapour pressure of the light actinide metals increases significantly in the series reaching a maximum at Am (Fig. 2) for which the vapour pressure is 0.1 kPa at 1500 K. This will complicate the fabrication and may lead to unwanted re-distribution during irradiation (as we will discuss below). Also the thermal conductivity of the actinide metals changes in the series and the values for the transuranium elements determined experimentally (Np, Pu) or predicted (Am, Cm) are lower than those of uranium. However, the values are still quite high compared to the corresponding ceramic oxides.

The properties of the minor actinide oxides are less well known than those of the metals [3], but the same trends are observed. The melting point, chemical stability and thermal conductivity all decrease along the dioxide series. For example, the oxygen potential above AmO_2 is very high, and CmO_2 even decomposes above 700 K. For these elements binary oxide with O/M lower than 2 must be considered, for example the sesquioxides An_2O_3. Their chemical stability is better and they melt at around 2500 K. Mixed oxides of the transuranium actinide thus will have a significantly lower melting point. Taking also into account the low thermal conductivity (Fig. 3), the margin to melting for mixed oxides with high concentrations of minor actinides will be significantly smaller than for UO_2, when operated with the same linear power. When mixed with an inert matrix such as ZrO_2 (melting point 2983 ± 15 K, but with a very low thermal conductivity [5]) this latter drawback becomes even more important, and will unavoidably lead to a limitation in the actinide content in the fuel or target. The same is true for mixed oxides with uranium oxide as matrix, though the effects are somewhat less prominent.
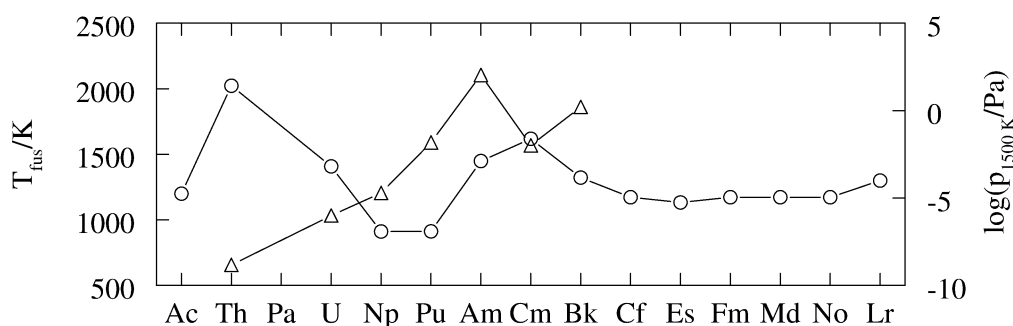

Figure 2. The variation of the melting point (○) and the vapour pressure at 1500 K (△) of the actinide metals.
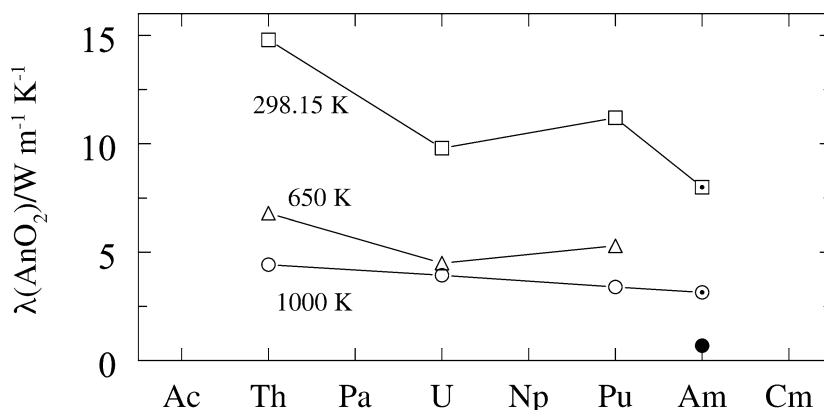
Figure 3. The thermal conductivity λ of the actinide dioxides. The experimental value (at 333 K) for AmO_2 is indicated by ●. This value is, however, suspicious and the measurement was probably made on a sub-stoichiometric sample. The estimated values for AmO_2 [17] are indicated by ⊙, ⊡.
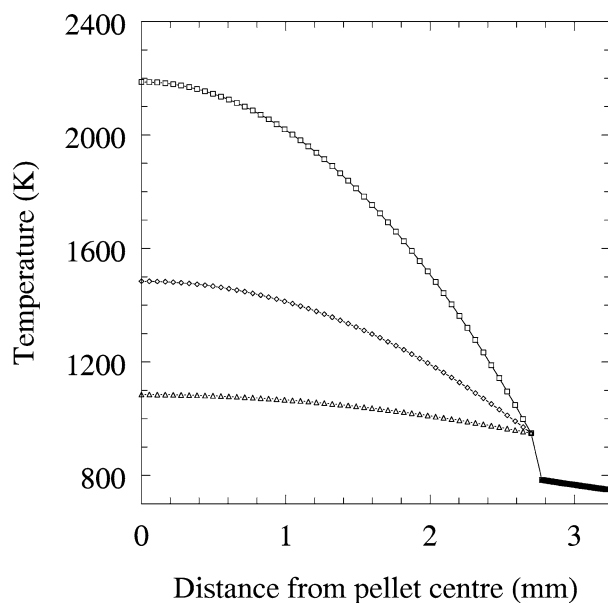
Figure 4. Fuel temperature profiles (beginning of life) for $(U, MA)O_2$ (□), a composite of this material with MgO (◊) and with Mo (△). The calculation was performed for a pellet with 5.4 mm diameter, a diametrical gap of 150 μm filled with He at 5 bar pressure at room temperature, a coolant temperature of 723 K, and a maximum linear heating rate of 280 $W \cdot cm^{-1}$.

The relevant properties of the actinide nitrides UN, NpN and PuN are fairly well established, those of AmN and CmN are unknown. Again a systematic decrease of melting point, chemical stability and the thermal conductivity along the actinide series UN–AmN is observed [4]. The thermal conductivity of the nitrides is high and, unlike the metals and oxides, increases with increasing temperature. The major concern is the thermal stability of the nitrides at high temperatures, as they decompose to nitrogen gas and the actinide metal, the latter in liquid and gaseous state. Especially for AmN this decomposition is expected to be significant, as was found experimentally for $(Pu, Am)N$ [6]. For uranium-free nitride fuels ZrN has been proposed as inert matrix, which is a stable and high-melting ceramic.

2.2. Composite fuels and targets

Some of the above noted drawbacks for homogeneous fuels and targets can be overcome by composite fuel designs. By the choice of an appropriate matrix, the overall properties can be tailored for specific applications. For example, by selecting a matrix with a high thermal conductivity the overall thermal conductivity of a heterogeneous mixture can help to reduce the operating temperature and thus increase the margin to melting (Fig. 4). This is especially relevant for oxide fuels. Ceramic and metal matrices are generally considered for ceramic-ceramic cercer or ceramic-metal cermet composites. For the cercer fuel types MgO is the main matrix candidate, for cermet fuel types a variety of metals are considered (Zr, Mo, W, Nb, stainless steel). Because the ceramic matrices are often sensitive to radiation damage from the fission products, which will have a negative effect on its properties, the optimum microstructure to limit the damage is provided by spherical particles homogeneously distributed in the matrix [7]. When the particle diameter is larger than about 50 μm and the volume fraction is of the spherical particles is less than 30%, the damaged volume fraction of the matrix is less than 10%.

3. Fabrication of MA fuels and targets

The radioactive characteristics of the relevant americium and curium isotopes are quite different from those of uranium and plutonium (see Table 1). The γ dose rates are orders of magnitude higher than uranium or even plutonium, which means that lead shielding is a prerequisite. In addition, the neutron dose rate due to spontaneous fission is very high for ^{244}Cm, which requires further shielding with materials with high hydrogen density like water or polyethylene. Finally the power produced by ^{244}Cm is significant and necessitates forced cooling when stored.

Thus the fabrication of MA fuels or targets can only be made in dedicated facilities. A typical example is the MA fuel fabrication chain that is being constructed at the Institute for Transuranium Elements (ITU) in Karlsruhe. The shielding of this chain is made of 500 mm water (for neutrons) and 50 mm lead (for γ radiation). Radiation dose calculations with a limit 2 μSv·h^{-1} at 1 m distance (independently for gammas and neutrons) gave a limiting mass of 5 g ^{244}Cm and 0.4 g ^{242m}Am for this facility. Significantly larger quantities of ^{237}Np, ^{241}Am and ^{243}Am can be handled. The limiting masses for these isotopes are determined by the operating license, however, and are determined by possible release of respirable powders (with aerodynamic diameters of $\leqslant 10$ μm (AMAD)) to the environment in the event of accidental conditions. The formation of respirable dust must also be avoided to limit the accumulation of material in the cells and thus the dose in case of eventual repair and maintenance of equipment therein. Especially for curium the effects on the fabrication processes is large [8]. The processes for the fabrication/handling of minor actinide fuels and targets must be as simple as possible and must be compatible with telemanipulation and extensive automation.

Table 1. Decay characteristics of some relevant actinide isotopes

Isotope	$t_{1/2}$	Activity	γ dose rate[a]	Spontaneous fission	Power
	y	Bq·g^{-1}	mSv·h^{-1}·g^{-1}	s^{-1}·g^{-1}	W·g^{-1}
^{235}U	7.04×10^8	8.00×10^4	1.3×10^{-3}	1.60×10^{-6}	6.00×10^{-8}
^{239}Pu	24100	2.29×10^9	1.5×10^{-2}	1.00×10^{-2}	1.93×10^{-3}
^{241}Am	433	1.26×10^{11}	312	0.48	1.14×10^{-1}
^{242m}Am	140	3.87×10^{11}	12	62	4.49×10^{-3}
^{243}Am	7370	7.40×10^9	44	0.27	6.43×10^{-3}
^{244}Cm	18.1	2.99×10^{12}	4.9	4.00×10^6	2.83

[a] at 1 m

Injection casting is the most common method for the fabrication of metal fuels. This process involves the melting of buttons of metal fuel alloys and casting the liquid in a mould to form fuel pellets. This process is in principle simple, dust-free and easy to automate, as has been demonstrated for the fuel fabrication of EBR-II [2]. However, when the MA content in the alloy is high, the vaporization of americium is unavoidable and its trapping and (highly efficient) recycling must be made.

Powder blending in combination with compacting and high temperature treatment is the most common fabrication technology for ceramic fuel pellets (oxides, nitrides). The process consists of many steps and especially during the blending/milling phase, needed to get a homogeneous material, dust formation is unavoidable. The use of free-flowing (i.e., dust free) powders as starting materials can help to overcome this. Such powders can be produced by liquid processing of the separated elements after reprocessing/partitioning, for example by sol-gel technology. For nitride fabrication, an additional problem arises from the very high temperatures needed for the conversion of oxides into nitrides by carbothermic reduction. During this process vaporisation of americium (metal) could occur. An additional difficulty of nitride fuel arises from the fact that the fabrication must be made with nitrogen enriched in ^{15}N ($>99\%$) to avoid ^{14}C production during irradiation. The nitrogen gas must be recycled effectively in the fabrication chain, as it is quite expensive [9].

A novel technology, developed at laboratory scale, is the infiltration of actinides into ceramic matrices [10,11]. In this process the highly active elements (Am, Cm) are infiltrated into porous pellets or powders (free flowing) by capillary forces. After the infiltration the precipitated actinides are converted to oxides by calcination. Next compaction, in case of powder infiltration, and sintering are applied. The major drawbacks of this method are the limited amount of actinides that can be incorporated into the matrix (about 30 wt% in ZrO_2) and the fact that the matrix should not be soluble in the slightly acid solution of the actinide to be infiltrated. The latter requirement limits the number of matrix materials and, moreover, excludes in many cases the possibility of hydrochemical reprocessing.

For the fabrication of composite fuels and targets a combination of the powder blending and infiltration methods can be used. Here one of the main technical difficulties is the realisation of a homogeneous distribution of the dispersed phase in the matrix, as generally significant differences in density exist between the matrix and the actinide phase. This will play an important role for industrial scale fabrication.

Further process simplification for ceramic fuels and targets could be achieved by selecting other fuel packing scenarios, for example, sphere-pac or vibro-pac fuel pins. In these processes the compaction step can be avoided and the sintered particles are loaded directly into the fuel pin and densified using a vibration method. Such a fuel or target will have a low smeared density (max 80–85% of the theoretical value) which has severe penalties on the thermal conductivity. Therefore vibro- or sphere-pac fuel designs for pure oxides are not likely. In that case a composite of oxide and metal spheres could be envisaged.

In all these cases, the fabrication process must permit recycling of the wastes and scraps from the fabrication process, to keep the losses per recycle steps as low as possible (see Fig. 1).

4. Helium build-up in MA fuels and targets

The experimental information on the irradiation behaviour of minor actinide fuels or targets is limited to the SUPERFACT and EFTTRA-T4 experiments. In the SUPERFACT experiment several uranium-based mixed oxides fuels containing neptunium and americium were irradiated in the Phénix fast reactor [12]. In the EFTTRA-T4 experiment a target of $MgAl_2O_4$ containing 12 wt% Am was irradiated in HFR-Petten [13]. Much has been learned from these experiments, especially in relation to the helium that is produced in the transmutation chain of americium (see Fig. 5). In the high Am content fuel pin of SUPERFACT experiment ($U_{0.6}Np_{0.2}Am_{0.2}O_{1.93}$) the helium production was about 25 times higher than in the Am-free pins. Although most of the helium was released from the fuel pellets during the irradiation, this led to a more than usual swelling of the fuel, interaction between fuel and cladding, and higher cladding stresses. The behaviour of the low Am content fuel pins of SUPERFACT ($U_{0.74}Pu_{0.24}Am_{0.02}O_{1.96}$) was close to that
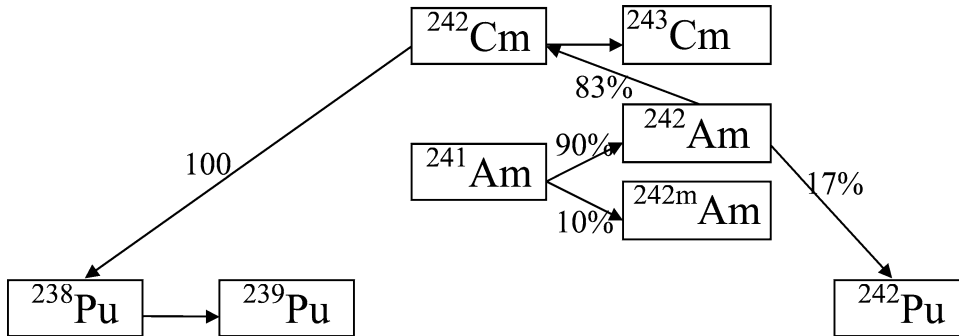
Figure 5. Transmutation scheme for ^{241}Am in a thermal flux. For the present context two points are of relevance: (i) because of the very short half life of ^{242}Am ($t_{1/2} = 16$ h) most of it will decay to ^{242}Cm, though its fission cross section is high ($\sigma_f = 2900$ barn), and (ii) the decay product ^{242}Cm has a low fission cross section ($\sigma_f < 5$ barn) and decays with a short half life of 163 days, producing ^{238}Pu and emitting an alpha particle.
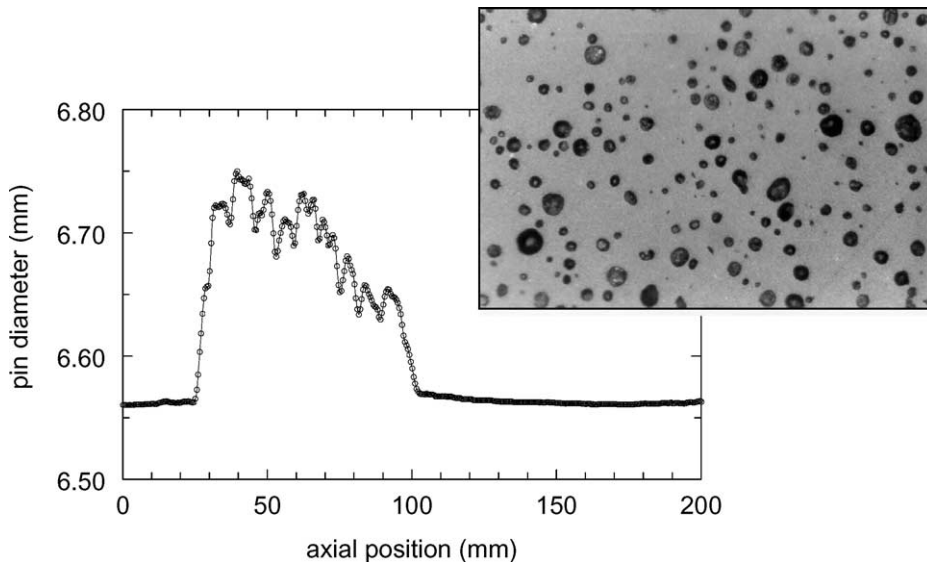

Figure 6. The external diameter of the EFTTRA-T4 fuel pin (original diameter 6.55 mm) showing the strong expansion due to the swelling of the pellets. The insert shows a micrography of the target revealing numerous gas bubbles formed though the accumulation of helium gas in the target. The measured increase in porosity (16%) corresponds well with the maximum volume swelling calculated (18%).

of standard oxide fuel under equivalent irradiation conditions [12]. Also the EFTTRA-T4 target swelled considerably due to helium formation [13]. In this target, which operated at relatively low temperature, the major part of the helium was accumulated in gas bubbles in the matrix, forcing the pellets to swell up to 18% in volume (Fig. 6).

It is generally accepted that the release of helium from transmutation fuel and targets is to be preferred, from the point of view of fuel behaviour during both normal operation and transient conditions. This is possible by several means:

(1) Anticipating pellet swelling by increasing the gap between cladding and pellet (therefore lowering the smeared density). The penalty of the large temperature increase in the gap early in life must be

compensated by liquid metal bonding. This concept is known from U metal fuel development in the USA where it was found that at a fuel swelling of about 30% the gas bubbles in the fuel start to interconnect to form paths for release [2]. Sodium bonding was used for this fuel type. Also for $(U, Pu)N$ fuel such a concept has been investigated successfully.

(2) High initial porosity so that a stable network of release paths already exists at the beginning of the irradiation. This has a large penalty on the thermal conductivity, where a composite design could provide a means to compensate this.

(3) Designed and controlled high operating temperature of the fuel to achieve thermally driven release from beginning of irradiation and minimal pellet swelling. The main penalty here is the smaller margin to melting, which is difficult to compensate. This concept was applied in the SUPERFACT fuels, for which the central temperature was around 2000 K.

Retention of helium, on the other hand, could be realised in coated particles in which the first porous buffer layer is designed to accommodate the helium plus fission gases produced. For fast reactor application the traditional TRISO concept (with layers of porous graphite, SiC and dense graphite) needs to be reconsidered.

5. Reprocessing of MA fuels and targets

As multiple recycling of MA fuels and targets is foreseen in most P&T scenarios, the possibility to reprocess the spent fuels and targets must be considered, and hydrochemical and pyrochemical techniques under discussion. The latter has the important advantage that the molten salt solvent is much more resistant toward radiation than the aqueous solvents. Also shorter cooling times before reprocessing can be applied.

For metal fuel only, pyrochemical reprocessing must be considered. The electrorefining process has been developed on a semi-industrial scale for the treatment of the metal fuel of the EBR-II reactor in USA. Tests to extend this process are being made in several laboratories around the world. The pyrochemical process must be performed in a very pure inert atmosphere, free from moisture and oxygen. The separated actinides can be collected in metallic form on a cathode and can then be used for production of new metal fuel.

Uranium oxide fuels are traditionally reprocessed by hydrochemical techniques. Also $(U, Np, Am)O_2$ can be dissolved in nitric acid, the reference solvent for hydrochemical processes, as has been demonstrated for the irradiated SUPERFACT fuels. However, with increasing Pu content in mixed oxide fuel, the dissolution kinetics in nitric acid decreases. When the fuels or targets are uranium-free the aqueous dissolution can become problematic; e.g., ZrO_2-based fuel is insoluble in nitric acid. The MgO matrix of a composite, in contrast, is easily soluble, but also the actinide phase must be soluble which will again be difficult if it is ZrO_2-based or rich in plutonium. Also pyrochemical reprocessing of such oxide fuels is not straightforward, as it needs a (high temperature) conversion step to transform the oxide into metal, which complicates the process.

Uranium-based nitride fuels are compatible with both hydro- and pyro-chemical processes. Also ZrN dissolves in nitric acid. The possibility to reprocess ZrN-base materials by pyrochemical methods is still under investigation. It has been suggested that the ^{14}C produced from the ^{14}N in the starting material should be recuperated in the reprocessing plant [14], which is easier with a pyrochemical process.

6. Discussion and conclusions

In the foregoing sections we have discussed in general terms the major technical problems that should be taken into account in the design of fuels and targets for transmutation. It is clear that any combination of the characteristics in Table 2, has positive and negative effects on the fuel cycle aspects and the selection of fuel and targets materials thus must be discussed in close relation to fuel cycle strategies.

In the 'single-stratum park' the reactors used for energy production and transmutation are the same and the transuranium actinides are recycled in the reactor fuel (small quantities) or in dedicated fuel assemblies (large(r) quantities). These scenarios are generally based upon incremental improvement of the existing

technologies and can therefore be described as evolutionary and their benefits are only evident in a long-term nuclear strategy [15]. Uranium-based metal or oxide fuels are the likely candidates, because for low minor actinide content (<5%) the properties of these fuel materials are changed only marginally. The development of such fuels can build on the experience from the US (metal) and the European (mixed oxide) fast reactor programmes. However, the metal fuel can only be used in liquid-metal and not in gas-cooled reactors because the margins to melting would be too small for the latter due to the higher coolant temperature.

The metal fuel has a better potential to cope with the more significant swelling caused by helium produced by transmutation of minor actinides, due to the possibility of increasing the pellet-cladding gap and the use of sodium bonding. Another important advantage of a metal fuel cycle is the possible de-centralised fabrication and reprocessing at the reactor site (co-location of facilities) because of the compactness of the installations and the integration of reprocessing and fabrication (no conversion steps are needed). However, a technically reliable solution for Am vaporisation during fabrication must be found.

The results of the SUPERFACT mixed oxide fuel pins with low Am content revealed no abnormal irradiation behaviour indicating the feasibility of this route. However, the number of subassemblies would be relatively high in case of such small minor actinide additions to the fuel. Therefore co-location would also be an advantage in this case to avoid long-distance transports of these active materials. This is, however, not likely when hydrochemical reprocessing of these fuels is made because the aqueous process is inherently more voluminous. The use of dedicated fuel assemblies with larger quantities of minor actinides is therefore to be preferred. The SUPERFACT experiment has shown that such a solution is feasible, although the irradiation behaviour at high(er) burn-up must be demonstrated. A special case is the so-called 'once-through-deep-burn' strategy with the aim to transmute more than 90% of the actinides in a single irradiation
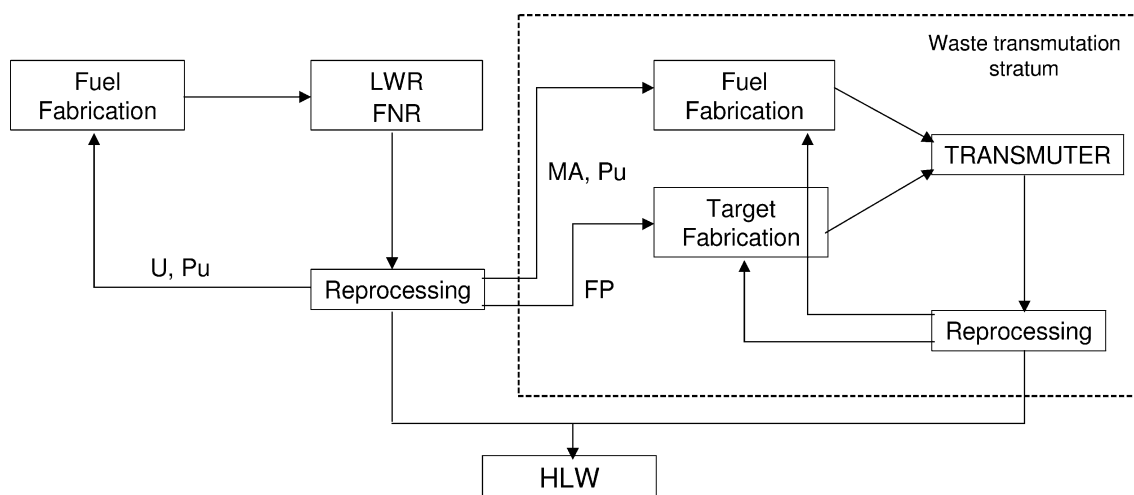

Figure 7. Schematic presentation of the dual stratum fuel cycle.

Table 2. Main characteristics of fuel and target forms

Nuclear character	fissile, fertile, U-free
Chemical state	Metal, oxide, nitride
Physical state	Single phase, homogeneous mixture (solid solution), composite
Packing	Compact, vibro- or sphere pac

followed by direct disposal of the spent material. In this case a fissile- or fertile-free oxide target must be used, for example ZrO_2-based [11].

In the 'dual stratum park' the transuranium actinides are transmuted in reactors dedicated to this task (Fig. 7). The fuel for the transmuter stratum will be optimised to achieve a maximum transmutation yield, which can mean radical designs compared to current technology: high (minor) actinide loading (20–40%) and high burn-up, preferably but not necessary in uranium-free targets. The major difficulty here are the many uncertainties in the design parameters for the fuel because such transmutation devices do not exist yet. Generally it can be concluded that the uranium-free metal or oxide fuel as homogeneous pellets are not very suitable: the metal because of unfavourable phase relations and Am vaporisation, the oxide because of the small margin to melting [16]. For the oxide this drawback can be overcome using composite fuels— as pellets, sphere pac or coated particles. This type of fuel will require extensive research to demonstrate its feasibility under the extreme conditions. Nitride fuels combine a number of advantages of metal (high thermal conductivity, sodium compatibility) and oxides (high melting point) fuels. However, very little is known about nitride fuel materials with high minor actinide content and extensive research is needed to demonstrate their technical feasibility, and if the ^{14}C problem can be managed.

In this context some words are needed concerning nuclear facilities and infrastructure required to develop transmutation fuels. The development of new fuels is a time-consuming task. It requires fabrication technology development, fundamental property measurements and irradiation testing over many years. As we have discussed, the facilities necessary for handling minor actinides are not standard and very few exist. For irradiation testing in representative (fast flux) conditions no facilities will be available in Europe after the closure of Phénix. Tests must therefore be made in materials testing reactors, but also their availability is uncertain in the longer term. Investment in facilities that are suited to handle minor actinides is a prerequisite for assessing the viability of Partitioning and Transmutation as a method to reduce the long-term radiological impact of spent nuclear fuel.

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