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Aspects of fabrication of curium-based fuels and targets

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Abstract

The limited fabrication possibilities will be decisive in the design of curium fuels and targets. It appears that there are significant advantages to be gained by separating the Am and Cm at the reprocessing stage, simply to concentrate the problems caused by Cm and limit its mass flow. Vibrocompaction of particles or spheres is probably the most suitable consolidation process, as it reduces the number of fabrication steps and reduces the concomitant controls and scraps occurring in pelletization routes for mixed fuel and targets. The infiltration process is the most promising method to convert aqueous Cm solutions to solid material, but sol–gel and other controlled precipitation for granulate production should not be neglected. Storage of the solidified Cm with a delayed introduction into the reactor for Plutonium transmutation is the most promising concept.

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1. Introduction

The different recycling modes for minor actinides will require the manufacture of innovative assemblies containing americium and curium. In presently considered scenarios [1], less than 5 wt% of Am and Cm oxides can be diluted in MOX or MIX fuel pins for homogeneous recycling in EFRs and PWRs, or larger quantities (up to 50%) of Am and Cm oxides dispersed in an inert matrix are considered for dedicated assemblies for heterogeneous recycling in EFRs or specialized reactors. In the latter case the inert matrix can be yttria stabilized zirconia (YSZ), magnesia (MgO), or a suitable metal (e.g. Cr, V, W, steel). The choice greatly influences the total flow rate in the fabrication plant and is close to a few hundred tons per year or a few tons in the homogeneous and heterogeneous recycling modes, respectively.

Manufacture of MA-assemblies is complicated by the alpha, beta, gamma and neutron emissions of americium and curium, whatever the MA content. Neutron activity and decay heat of curium is especially penalizing compared to standard MOX and UO₂ fuels. Thus the use of

conventional processes and technology based on powder metallurgy is questionable.

To illustrate the fabrication issues due essentially to the curium, this paper investigates the consequences of the curium neutron and thermal radiation on the process and facilities for fabrication of advanced composite fuels or targets, as designed in the current R&D studies [2,3]. It will be shown that, in contrast to conventional UO₂ or MOX fuels, *the limited fabrication possibilities will be decisive in the design of the fuel*. Some recommendation on fabrication process and consequently on MA-fuel microstructure will be given.

2. The curium issue: high neutron source and decay heat

Radiological characteristics of MA-assemblies and fabrication facilities are recalled in Table 1. They have been assessed for three different scenarios, listed below:

1. Once-through recycling (heterogeneous mode) of Am and Cm in a mixed park of PWRs and EFRs. Am and Cm are transmuted in dedicated assemblies, moderated with YH_x [4] and loaded only in the EFR fleet.

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Table 1
Activity, decay heat and neutron source at the fabrication stage

			Values per ton of heavy metal atoms		Values per assembly	
			Standard fuel ^a	Transmutation fuel	Standard fuel ^a	Transmutation fuel
1	Once-through heterogeneous recycling in EFRs ^b	Activity (Bq)	4.1×10^{16}	2.9×10^{17}	4.4×10^{15}	2.5×10^{15}
		Neutron source (n/s)	2.5×10^8	7.2×10^{11}	2.7×10^7	6.3×10^9
		Decay heat (kW)	5.3	270	0.8	2.3
2	Homogeneous multi-recycling in EFRs	Activity (Bq)	4.1×10^{16}	3.6×10^{16}	4.4×10^{15}	3.8×10^{15}
		Neutron source (n/s)	2.5×10^8	1.4×10^{11}	2.7×10^7	1.5×10^{10}
		Decay heat (kW)	5.3	8.4	0.8	0.9
3	Homogeneous multi-recycling in PWRs	Activity ^c (Bq)	5.7×10^{16}	2.1×10^{16}	3.0×10^{16}	1.1×10^{16}
		Neutron source (n/s) ^c	1.4×10^8	2.9×10^{10}	7.3×10^7	1.5×10^{10}
		Decay heat ^c (kW)	3.1	9.4	1.7	4.9

^a An EFR assembly with (U, Pu, Np)O₂ containing 20% Pu and 5% of Np for fast reactors (107 kg HM), or a PWR assembly with (U, Pu)O₂ fuel containing 12% of Pu for PWRs (520 kg HM).

^b The Am and Cm targets contain 8.7 kg HM.

^c Including the ²⁵²Cf not separated from Am and Cm by aqueous processes.

- Am, Cm and Np are diluted in the driver fuel of the EFRs (homogeneous mode). The nuclear park is equipped only with EFRs.
- Am, Cm and Np are diluted in the MIX fuel (enriched in plutonium and uranium 235) of PWRs (homogeneous mode). The nuclear park is equipped only with PWRs.

In all cases, a nuclear park with a total power of 60 GWe supplying 400 TWeh per year is considered. The data are presented in Table 1 for the equilibrium state, i.e. when the actinide production equals the actinide consumption.

Neutron and other radiation sources require appropriate radioprotection measures, i.e. remote handling in heavily shielded cells. This implies a large financial investment and higher maintenance costs, than found for example for MOX fuel, will ensue. To keep the fabrication step as competitive as possible, *these drawbacks should be balanced by process robustness and facility compaction.*

Furthermore, the heat released by curium (2.8 W g^{-1}) is 28 times higher than that released by americium, and leads to new technical problems to be mastered everywhere in the plant. One liter containing 250 g of actinide, a concentration often required in some liquid to solid conversion processes [5], releases about 25 W for Am, about 100 W for an Am/Cm ratio of 88:12 (representative of scenario 1 above), and 700 W for Cm alone. This should be compared to the highest thermal power of 10 W/l encountered at the La Hague plant fission product concentrate tanks. In the same way, to keep 1 m² of a Am + Cm powder bed at a temperature below 80 °C, the maximal thickness of the bed must not exceed 0.5 mm to favour natural cooling by air. Otherwise forced cooling

is necessary. *No industrial experience is available today to manage such solution or solid batches.*

3. Impact of curium on the fabrication process of MA-fuels and targets

3.1. Design of advanced fuels and targets

There are many features to consider in the design and qualification of fuels bearing minor actinides, e.g. the irradiation behaviour of the different materials (inert matrix, moderator and actinide compounds), the helium production, etc. Currently two types of microstructure are favoured for MA-based targets and fuels [2,3]:

- a homogeneous refractory material (CER), or
- a heterogeneous composite of a MA-based compound diluted in an inert ceramic or metallic material (i.e. CERCER or CERMET, respectively).

Yttria stabilized zirconia has a cubic structure and is the (currently) preferred CER matrix. Its main disadvantage, however, is its low thermal conductivity, which could limit the maximum MA content (2.2 g cm^{-3} being required in scenario 1 [5]) in order to maintain a sufficient margin between the melting point of the fuel and its operating temperature. This limitation can be overcome by incorporating higher concentrations of MA in YSZ particles, which are then diluted in another material with a higher thermal conductivity such as magnesia or steel to give a CERCER and CERMET, respectively. In the limiting case, the YSZ could be removed and the composite material could be based on (Am, Cm)O₂ particles. The preferable MA-particle form is a sphere

with a diameter of at least 100 μm , so that damage, and concomitant property deterioration of the surrounding diluting matrix during irradiation is minimized. That requires the production of fuels and targets with a good size and spatial distribution of the spheres throughout the inert matrix.

Fuel consolidation is achieved traditionally by pelletization and sintering of the homogeneous or heterogeneous powders. Here a controlled gap between the pellet and the cladding and/or an adequate stable open porosity must be available to enable fuel swelling and gas release from the fuel to the pin plenum. This is particularly important for MA's due to the release of helium during the irradiation. Manufacture of such advanced fuels, meeting all the requirements explained above, leads to many process and control steps, which contribute not only to the complexity of the process but also to increase scraps, and primary or secondary waste volumes.

3.2. Impact of curium on the fabrication process

Technological constraints linked to curium handling are particularly penalizing for the following steps:

Conversion step: Powder metallurgy must be avoided as the generated fine and disperse dust is unacceptable. Therefore, direct synthesis of spherical dust free particles containing the MA's is required. Thus, if the installations can remain clean, operator intervention can be considered, provided all production material is removed from the chain in advance. This hybrid concept is being tested at the ITU, albeit for laboratory scale quantities (100 g Am and 5 g Cm). If it is not successful, maintenance must be made by remote handling, as is the case for the vitrification facility at the La Hague plant. Auxiliary units needed for the maintenance (handling crane, special cells, etc.) would have to be integrated in hot cells also.

Powder pressing: At an industrial scale, automatic pressing requires the use of lubricants, mixed generally to the powder just before the pressing step. In MA fuels they could lose their lubrication properties rather quickly. Further degradation leading to mechanical instability of green pellets during storage is expected too. Such a phenomenon has been noticed during the fabri-

cation campaign of the CAPRA assemblies [6], containing degraded plutonium with a high ^{238}Pu content. That means that development of automatic lubrication of press dies would be necessary to compact MA-pellets.

Assembly mounting and storage: Assembly mounting would also have to be performed remotely behind concrete shielding walls, a step further complicated by the large dimensions involved (4.8 m for an EFR-type assembly). Forced cooling would be also necessary to keep the temperature of the structural materials at a reasonable temperature to avoid their damage, during handling and storage.

In conclusion, neutron activity and gamma power of curium necessitate thick concrete or water shields and continuous forced cooling everywhere in the fabrication facility from the conversion step to the assembly mounting, and also during its transport and handling in the reactor facility. Process compaction to limit cell volumes and number, and simplification to facilitate the equipment maintenance and to reduce as much as possible the volume of liquid and solid waste contaminated with curium are therefore of the greatest importance.

In contrast to UO_2 or MOX fuel, fuel design sophistication to increase performance in pile is a secondary priority. The main objective is the design of simple, robust and compact fabrication routes to limit fuel controls and scraps.

4. Recommendations for Cm fabrication processes

Different routes should be studied in-depth with the aim to simplify and compact the fabrication process. Some of them are described below.

4.1. Reduction of the Cm-based mass flow by separating Cm from Am

Table 2 gives the actinide mass flow in different P&T scenarios. It highlights the interest of the heterogeneous recycling in standard reactors (case 1) or the recycling in reactors dedicated to the transmutation (case 4) since the curium mass flow in the fabrication plant and number of MA-assemblies is dramatically reduced. The separation of Am from Cm is especially interesting as the flow of

Table 2
Mass flow of heavy atoms in the fabrication plant

		Mass flow of heavy metals (t/y)					Total
		Cm	Am	Pu	Np	U	
1	Targets with Am and Cm	0.2	1.2	0	0	0	1.6
2	EFR fuels with Am, Cm, Np	0.6	2.3	57.6	0.3	295	356
3	PWR fuel with Am, Cm, Np	3.6	2.6	23.2	1.0	787	817
4	ADS fuel with Am and Cm	1.1	2.4	2.1	0.0	0.2	6

Table 3
Impact of Cm separation on the fabrication facility (case 1)

	Am + Cm-targets	Am-targets ^a	Cm-targets ^a
Isotopic composition	²⁴⁰ Pu:0.8/ ²⁴¹ Am:57.8/ ^{242m} Am:1.2/ ²⁴³ Am:28.8/ ²⁴³ Cm:0.2/ ²⁴⁴ Cm:9.8/ ²⁴⁵ Cm:1.1	²⁴⁰ Pu:0.5 ²⁴¹ Am:76.3% ^{242m} Am:1.7% ²⁴³ Am:21.3%	²⁴³ Cm:3.1% ²⁴⁴ Cm:86.8% ²⁴⁵ Cm:9.5% ²⁴⁶ Cm:0.5%
Mass (tonnes pa)	1.71	1.51	0.2
Number of pins	56 160	49 263	6897
Number of assemblies	180	158	22
Activity (Bq/ <i>t</i> _{HM})	2.9 × 10 ¹⁷	1.1 × 10 ¹⁷	2.7 × 10 ¹⁸
Heat decay (kW/ <i>t</i> _{HM})	270	90	2580
Neutron source (n/s/ <i>t</i> _{HM})	7.2 × 10 ¹¹	2.8 × 10 ⁹	1.0 × 10 ¹³

Including ≈0.8% decay to ²⁴⁰Pu.

^a Assuming that the Am or Cm pin and assembly design is similar to that of (Am + Cm)-target.

Cm-containing product is reduced from 1.6 t to only 200 kg per year and decreases the number of assemblies from 180 to 20 per year (see the example of case 1 in Table 3). The mass flow reduction is accompanied by thermal and neutron source increases of 30 and 3500, respectively. In contrast, the technological constraints on the Am fabrication line are relaxed. Here the shielding is predominantly lead, though neutron shielding against (α,n) reactions is required. A hybrid type cell with operator maintenance should be achievable.

4.2. Suppression of dry powder metallurgy steps (Vibrocompaction)

Vibropacking (VIPAC) is an alternative process to pellet pressing to consolidate fuel or target particles. The pins are loaded directly with granulates or microspheres and compacted by gentle vibration to reach the target smear density. Smear densities of 85% can be usually achieved by selecting appropriately sized particle fractions, and the remaining open porosity provides a path for helium release to the fuel pin plenum.

This process has been used in Russia for (U, Pu)O₂ fuels following Pu and U recovery by pyrochemistry (VIPAC process [7]). It is well suited to fast neutron reactors since a high operating temperature is required for in-pile sintering of the particles. Tentative development has been made also in Europe for sol-gel microspheres (SPHEREPAC) in nitride and carbide fuels, but was stopped prematurely [8].

In addition to the suppression of dirty operations (granulation, pressing), the VIPAC/SPHEREPAC process has several advantages over pelletization. In particular pelletization introduces a series of additional specifications to be controlled and verified, e.g. density, diameter, height, visual aspect, microstructure, homogeneity, etc.. All of these are eliminated with VIPAC or SPHEREPAC consolidation and replaced by less intensive control on welded pins, i.e. actinide and density distribution. These controls are considerably easier and

Table 4
VIPAC/SPHEREPAC particle size required for a smear density of 85%

Particle size (μm)	vol.%
1200	60
300	15
30	25

cleaner to perform. In addition, the suppression of controls automatically decreases process scraps.

Application of VIPAC/SPHEREPAC to homogeneous fuel like (Zr,Y,MA)O₂ should be possible provided the microspheres or granulates of suitable composition and different sizes are easily synthesized. Usually three particle sizes are used (see Table 4). In the case of heterogeneous fuel such as MgO + (Am, Cm)O₂ or (Zr, Y, MA)O₂, some complications arise due to density differences, as no segregation is allowed. To reach an actinide density of 2.2 g cm⁻³, the large particles can be (Zr,Y,MA)O₂ and the medium and small sized fractions MgO. The MA content in the large spheres should be increased to about 3.3 g cm⁻³ to maintain an overall density of 2.2 g cm⁻³ in the pin. Conversely, the larger (and smaller) size fraction particles can be MgO and the medium size particles (Am,Cm)O₂.

4.3. Preparation of particles for vibrocompaction

The preparation of particles directly from the Am and/or Cm solution is the key step in the VIPAC or SPHEREPAC process. The liquid–solid conversion phase should allow the direct fabrication of the granulates or microspheres with the targeted composition and size, but could also include the co-precipitation of the inert matrix to avoid the segregation issue in the vibropacking step. Different routes could be considered to synthesize the particles to be loaded directly in the pins. In aqueous media they are:

1. co-precipitation or gelation via hydroxide route,
2. co-precipitation, via oxalate, formate or other organo-metallic compounds,
3. the infiltration process,
and in molten alkali chloride melts:
4. fractional precipitation of oxide (Am, Cm),
5. preliminary electrodeposition of metal (Am, Cm) in liquid metallic cathode followed by the removal of the metallic solvent and an oxidation step.

4.3.1. Aqueous co-conversion processes including sol–gel

The synthesis of granulated powder by direct co-conversion of Am and/or Cm aqueous solutions requires research and development. In particular, the behaviour of concentrated solutions containing curium and/or americium and the consequences on the co-precipitation of actinides must be investigated. The stability of the oxidation states of the actinides to control the composition and the main physico-chemical properties of the precipitate as final or intermediate product needs the following investigations:

- thermodynamics and kinetics of reactions involving actinides in concentrated solutions,
- radiolysis and thermolysis consequences on the stability of the species in solution
- stability of additives and the influence of impurities and unstable phases on the co-conversion efficiency.

Co-precipitation of actinides either by ammonia route (hydroxide) or by carbonate and carboxylate routes (formate, oxalate, ...) could be promising as low concentration solutions (e.g. 20 g actinides/l) with lower heat production are generally used with excellent precipitation yields. Some progress have already been made for curium and americium precipitation at a laboratory scale [9]. The exact structure of these Cm-based compounds are not precisely known, especially when curium is not the only actinide involved (mixed structure). Nevertheless, the main challenge is to produce large (100 μm) cohesive particles, which do not produce dust. Final thermal treatments need careful control to obtain the final product with precise composition (O/M) and phase structure. Indeed many are not fully characterized yet.

The sol–gel process is particularly adapted to the production of microspheres either by internal or external gelation processes. In the internal gelation process droplets of the feed solution, containing only actinide(s), or actinide(s) with zirconia, are internally hydrolysed by the means of chemical agents introduced in the solution, which thermally decomposed to produce ammonia. The thermal decomposition can be induced in a hot silicon bath, where the droplets are collected, or in a microwave furnace. In the external gelation process, droplets containing the actinide and organic polymers are collected

in an ammonia bath to be hydrolysed by diffusion of ammonia from the surface to the centre of the droplet. The particle size can be selected by the type of liquid dispersion device. Vibrating nozzles produce relatively monodisperse particles, while polydisperse particles are produced by rotating cup atomizers. The choice depends on the required fuel form, with the former preferable for SPHEREPAC and the latter for pelletization of homogeneous fuels.

The external gelation process has been used successfully for the production of (U,Np,Am) O_2 pellets for the SUPERFACT irradiation experiment [10]. In principle it could be used for the liquid to solid conversion of Cm containing solutions, but rapid processing following polymer addition is required to avoid their degradation by the a particles [11]. The concentrated (≈ 250 g/l) stock solution and the sol gel feed solution (100 g/l) would require cooling to remove excess heat.

Rather than use the sol–gel process to generate (Am,Cm) O_{2-x} , a CER fuel can be generated by diluting the minor actinides, as appropriate, in a Zr/Y solution. Although the MA flow remains constant, the mass flow increases substantially. This dilution in zirconia reduces the heat removal difficulties.

Perhaps the overriding difficulty in the conversion of Cm containing solutions to solid by any precipitation or sol–gel step is the handling of the liquid wastes, derived either from washing the gelled particles or from filtrating and rinsing the precipitate cake. The handling of such liquid wastes containing albeit small quantities of materials with high activity could rule out sol–gel or precipitation conversion steps for Cm. Sol–gel processing of solutions containing Am alone as minor actinide could still be feasible.

4.3.2. Infiltration process

The infiltration method (see Fig. 1) more than all others is worthy of further development [12]. In particular the process steps are simpler and *no liquid waste is produced*. The process requires a porous medium into which the Am + Cm nitrate can be infiltrated. In the EFFTRA-T4 experiment [13], for example, the porous medium was a magnesium aluminate spinel pellet formed by compaction of a commercial powder and subsequent calcination to remove the lubricant (zinc stearate). The low porosity of the pellet renders this method unsuitable for the production of targets with minor actinide contents of 2.2 g/cm³. Achieving a homogeneous Am distribution in a single infiltration step was already difficult for pellet infiltration, so that multiple infiltration/calcination cycles to increase the actinide content are unlikely to be successful.

In contrast to pellets, porous beads made by the sol–gel external gelation method have a much higher porosity. Infiltration can be made in a batch mode with addition of the minor actinide solution to the beads until

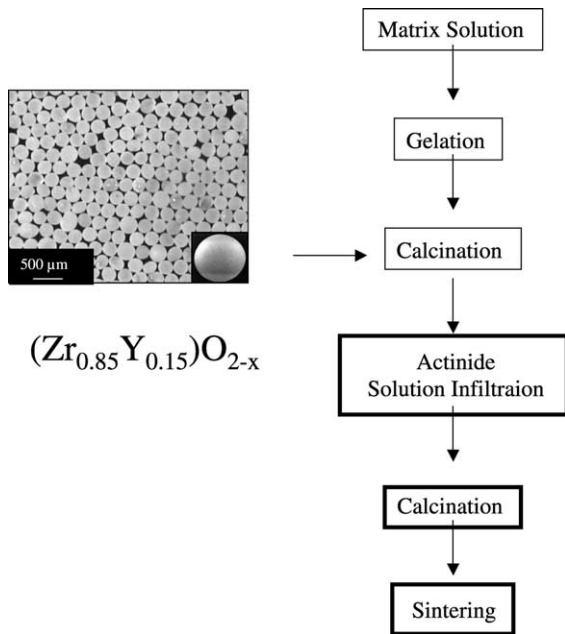


Fig. 1. The infiltration process for the production of SPHE-REPAC fuel.

the incipient wetness point is reached. In a single infiltration with a 200 g/l plutonium solution, 11 wt% Pu has been infiltrated into $(\text{Zr},\text{Y})\text{O}_2$ beads [14]. The relatively small size of the beads (20–120 μm) yields homogeneous products. Using multiple infiltration/calcination cycles Pu densities of 2.2 g cm^{-3} have been obtained in YSZ.

Adaptation of the process to separated Cm solutions is possible but the heat generation (560 W/l in a 200 g/l feed solution) must be managed. Process improvements are still possible. The porosity of the beads could be increased still further. Innovations to continuously infiltrate porous beads are under consideration.

If the porous beads are an inert matrix such as YSZ, they can be manufactured by the sol-gel external gelation method in conventional laboratory installations, where wastes can be handled, treated or recycled easily. Were it deemed necessary, the porous beads could also be actinide oxides, as long as they are insoluble in weakly acidic solutions. Regardless of the porous material, the infiltration of Cm solutions yields *de facto* no liquid wastes, an advantage of decisive importance in the selection of the fabrication process.

In contrast to the sol-gel process, the concentration of MA that can be infiltrated into a porous medium is limited. For CER fuels such as $(\text{Zr},\text{Y},\text{MA})\text{O}_2$, a minor actinide density of 2 g cm^{-3} can be achieved, but a CERCER of $(\text{Zr},\text{Y},\text{MA})\text{O}_2$ dispersed in MgO would require an actinide content in the zirconia phase, which is not presently achievable using the infiltration method.

Some difficulties linked to the curium presence, like the viscosity change of the solution, the thermolysis and radiolysis effect in solution or at the solid/liquid interface deserve special attention in the future development of this technique.

4.3.3. Molten salt processes

Molten alkali chlorides are ionic liquids, which are not subject to radiation influence. In such melts, neutron moderators are absent, therefore problems of criticality are solved more easily. Both factors permit working with high radiation loads and with high concentration of fissile materials.

Processes in molten alkali chlorides have been developed by RIAR for production of oxide (U,Pu) granulated fuel using oxide electrodeposition [15]. In normal conditions americium and curium practically do not co-precipitate into the cathode deposit. One possible way for recovering them as oxide involves fractional precipitation by adding sodium carbonate. Such a separation technique is investigated in the frame of a collaboration between RIAR, ITU and CEA. Preliminary experiments have demonstrated that the extraction of americium oxide is achievable in NaCl–KCl and NaCl–2CsCl melts. Laboratory research is required, however, to optimize the recovery of Am (and then Cm) and to assess their separation efficiencies from troublesome fission products (rare earth elements).

If the oxide precipitation is not effective (Cm behavior is currently unknown), an alternative route could include the recovery of Am and Cm as metal by co-electrodeposition in a liquid metallic cathode in a LiCl–KCl melt. This should be feasible since the reduction potential of An(III)/An(0) in LiCl–KCl are close using liquid cathodes (as has been proved for Pu, Np and Am with Cadmium) [16]. Similar electrochemical behavior can be expected with Cm [17].

If a Cadmium-based cathode is used, it can be separated from minor actinides by distillation: the metallic residue (Am–Cm alloy) is then oxidized for oxide production. The assessment of the americium recovery in liquid cadmium cathode is underway in many labs thorough the world [16,18] but must be extended to Cm.

5. Transmutation or intermediate storage of curium

The fabrication constraints could be further relaxed if Cm-based elements are not transmuted in pile but stored till complete decay into plutonium occurs. For example, porous YSZ beads could be infiltrated with Cm solution to the desired content, sintered and then directly poured into vessels, especially designed for interim storage. On a time scale of 100 years engineering barriers to Cm or Pu release should be sustainable.

Following storage, the Cm containing particles will have accumulated considerable quantities of He due to α decay. In addition, the matrix will be damaged extensively by the same process. Thus the particles should be heated to remove He and cure radiation damage, before being loaded into SPHEREPAC pins for transmutation of the produced Pu daughter products. The mode of transmutation (once through and then out, or multiple recycling) will depend on the total Pu inventory to be transmuted. This scenario is not the subject of the present paper, but its application is worthy of a detailed case study.

6. Conclusions

Apart from the shielding requirements necessary for the fabrication of fuels and targets containing curium, various processes would require additional engineering innovation for simplification and automation. Many of these are already available, or at an advanced stage, at MOX fuel fabrication plants, but would need to be adapted further to reduce maintenance, operator intervention, and to provide extra protection for electronic equipment.

The location of the reactor, reprocessing and fabrication sites also needs to be considered. In order to avoid difficult (trans)national transports, the location of all such facilities at one site would seem preferable. If not possible, there are considerable advantages in the co-location of reprocessing and fabrication facilities for Cm fuel and target production at the same site, as the minor actinides are in liquid form at the end of aqueous reprocessing streams.

As shown in Table 3, the number of assemblies containing Cm is reduced if it is separated from Am in the reprocessing steps. It would also appear advisable that Cm transmutation assemblies contain as many Cm pins as possible for ease (or at least concentrating the difficulties) of handling. Mixing of Cm pins in either UOX or MOX assemblies does not seem a suitable option, and should be avoided. Reduction of the mass flow achieved by the separation of Cm from Am cannot be overlooked, even if it leads to forced cooling of highly concentrated Cm solutions.

Infiltration of Cm solutions in YSZ or even AmO₂ seems viable and has the distinct advantage that no liquid wastes are produced. The microstructure of MA-targets will be determined by the limited fabrication possibilities. Direct vibropacking of these particles into pins should be one of the best consolidation methods, as it reduces the number of fabrication steps, removes several control steps, and eliminates a source of non-conforming materials. Maintaining homogeneity, if materials of different density are used, is a challenge to be mastered. Ease of fabrication must be balanced, however, with acceptable performance in-pile.

7. Recommendations for future investigations

New processes based on direct synthesis of granulates or green bodies from actinide solution must be investigated. Co-conversion processes, via oxalate, hydroxide or formate phases, could be studied to generate flowable and dust free particles of Am and/or Cm oxides of suitable composition and size. Though sol-gel processing is questionable, improvements are still possible and should be investigated before discarding such a method. Waste treatment could be alleviated if an oxalate rather than ammonium hydroxide step were used.

The infiltration method requires research and development to develop highly porous YSZ (and eventually AmO₂) spheres with dimensions suitable for SPHEREPAC fractions or pelletization and to improve the infiltration process to permit continuous infiltration of dilute rather than concentrated solutions.

Finally, the separation of Cm and its solidification (by the infiltration technique) in yttria stabilized zirconia microspheres, and its subsequent controlled storage limits the number of steps in which Cm has to be handled. Following the decay of the majority of the Cm to Pu, all subsequent steps to fabricate the fuel can be made without excessive shielding and the Pu can be incinerated in dedicated assemblies. The consequences of this option on the fuel cycle needs further assessment.

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