

**IMPACT OF THE CURIUM MANAGEMENT ON THE FABRICATION OF MA-BEARING
TARGETS AT AN INDUSTRIAL SCALE IN THE FRAME OF A MIXED PWR
AND FR P&T SCENARIO**

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Abstract

From the detailed analysis of the once-through transmutation scenario of americium and curium in moderated target assemblies in sodium-cooled fast reactors, it has been underlined the large technological impact of the curium management on the target cycle and especially on the fabrication process and plant. Because of the high heat decay and neutron emission of curium, any dry or wet fabrication process will require remote handling and continuous cooling and will be set up in hot cells, behind thick concrete shields, from the conversion to the assemblies mounting units and during transport. The target fabrication facility should look like to the La Hague vitrification plant. That means that one of the priorities in the further R&D programme on P&T should be to develop simple, compact and robust fabrication processes for TRU fuels and targets. VIPAC and SPHEREPAC processes are thought to be promising.

Introduction

The once-through heterogeneous transmutation of americium and curium (MAs) in sodium-cooled fast reactors, like EFRs, implies to design innovative assemblies (named target assemblies) compared to standard fuel assemblies. They are made up of pins filled with a mixing of americium and curium oxides and inert matrix (magnesia) and contain a hydride-based moderator (YH_x) to reach the minimum MAs fission rate of 90% requested to dispose of the target assemblies directly in a repository without reprocessing.

Fabrication of such targets is complicated by the alpha, beta, gamma and neutron sources due to americium and curium. Neutron activity and decay heat of curium is especially penalising compared to a standard fuel as shown in Table 1.

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

	Values per tonne of heavy atoms		Values per assembly	
	Standard EFR-type fuel	MAs-target	Standard EFR-type fuel	MA-target
Activity (Bq)	$4.1 \cdot 10^{16}$	$2.9 \cdot 10^{17}$	$4.4 \cdot 10^{15}$	$2.5 \cdot 10^{15}$
Neutron source (n/s)	$2.5 \cdot 10^8$	$7,2 \cdot 10^{11}$	$2.7 \cdot 10^7$	$6.3 \cdot 10^9$
Decay heat (kW)	5.3	270	0.8	2.3

The objective of this paper is to assess the consequences and the impact of the curium radiation and decay heat on the fabrication process and facility.

Neutron and radiation sources can be controlled by taking the appropriate radioprotection measures and by remote handling in hot cells. That means however heavy investment and maintenance costs which should be balanced by process simplification and compaction and increase of tools robustness. The challenge is nevertheless more economical than technical.

On the contrary, the heat released essentially by curium, poses a new technical problem since thermal power is a parameter to be controlled everywhere in the plant and during the targets transport to avoid tools damage, target melting or solution boiling according to the process.

Criticality has not been studied in-depth: rough evaluation does not reveal any difficulties.

All these constraints force to avoid transport at each step of the process and to militate in favour of the integrated cycle, that is the co-location of the reprocessing and re-fabrication facilities on a same site.

Characteristics of target assemblies to be fabricated

Different design of moderated target assemblies have been studied in order to optimise the transmutation capacity of the target while meeting thermal, thermo-mechanical and hydraulics requirements. A heterogeneous assembly, where moderator and MAs are distributed in separate pins, has finally been selected. Transmutation performance should reach the required values of 100-110 g of fissioned MAs per TWeh of EFR [1] and the technical feasibility should be guaranteed provided that some convenient optimisation studies are carried out on the behaviour under irradiation of moderator and MAs-based composite materials.

MAs content of the target pin, selection of hydride compound, moderator and MAs pin design have been optimised on the basis of the current knowledge about inert matrix fuels and americium targets coming from EFFTRA, CAPRA/SPIN and IMF programmes. [2]

Microstructure of MAs composite is optimised: (Am, Cm)O_{2-x} is homogeneously dispersed in magnesia under the form of spherical particles of 100 µm to limit the matrix damaging by fission products. Gap between (Am, Cm)O_{2-x} pellets and clad is controlled to accommodate a pellet swelling rate of 10 vol%. The maximum central temperature of the target pin is limited to 2 060°C to keep some margins with regard to the (Am, Cm)O_{2-x} melting.

Table 2. **Characteristics of the target assembly**

Assembly	
Internal across flats distance	174.2 mm
Pins number	
MAs pins	312
Moderator pins	157
MAs pins	
MAs isotopic composition (wt%)	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
(Am, Cm)O ₂ content	20.7 vol%
(Am, Cm)O ₂ particles size	100 µm
Fuel column length	1 000 mm
Outer pin diameter	5.23 mm
Outer pellet diameter	4.4 mm
Clad thickness (EM12 steel)	680 µm
Spacer wire diameter	732 µm
Spacer wire helicoidal pitch	180 mm
Moderator pins	
Hydride composition	YH _x , with x≥1.75
Outer pellet diameter	7.74 mm
Outer pin diameter	8.46 mm
Clad thickness (EM12 steel)*	300 µm

* the internal clad surface is coated with a molybdenum or tungsten layer of 10 µm of thickness to limit the hydrogen permeation from the hydride thermal dissociation .

Concerning the moderator, yttrium hydride has been preferred to calcium or zirconium hydride because of the best compromise between thermal stability and hydrogen content. It is expected that less than 10% of hydrogen would be lost by the moderator pin for a maximum operating temperature of 546°C. It should be possible to increase the operating temperature of YH_x up to 700°C by coating the internal surface of the clad with a molybdenum or tungsten layer, acting as a hydrogen permeation barrier.

Cladding material for MAs and moderator pins is the ferritic-martensitic EM12 steel which should withstand the high damage dose of 200 dpa NRTFe.

Finally, the heterogeneous assembly is based on the CAPRA design [3] with 469 pins. Table 2 summarises the main characteristics of the target assembly and Figure 1 gives a schematic view of the optimised assembly.

Fabrication of target assembly at an industrial scale

Production capacity and flow sheet of the fabrication facility

The fabrication process starts after the reprocessing and partitioning of irradiated fuels. The fabrication process input is then a solution containing 0,35 to 0,45 g of americium and curium per litre. The output is the MA-assembly, ready to be transported and loaded in fast reactor.

The fabrication process is composed of five units:

- the concentration unit: the diluted solution is concentrated before feeding the sol-gel process;
- the conversion or sol-gel unit: solution of americium and curium nitrate is converted into mixed americium and curium oxide under the form of spherical particles of 100 µm;
- the pellets and pins fabrication unit: pellets are pressed, sintered and loaded in pins. Pins are then welded;
- the assembly mounting unit;
- the waste and scraps recycling unit.

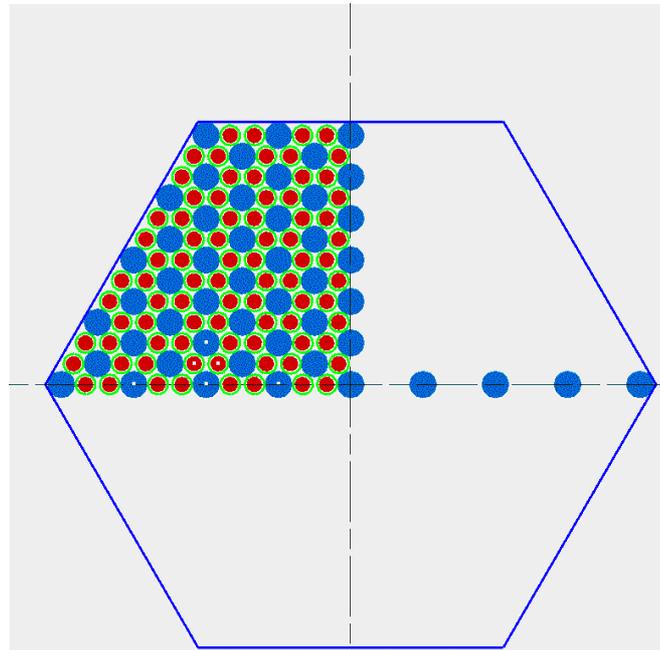
It is assumed that the non-radioactive moderator pins are fabricated outside the target fabrication plant and are delivered ready for the assembly mounting.

The annual MAs mass flow in the fabrication plant has been assessed to 1.51 t of americium and 0.2 t of curium. [4] 56 160 target pins and 180 target assemblies must be fabricated per year.

The neutron activity of curium rules out the possibility of setting up any part of the process in glove-boxes. It is necessary to install all the active process equipment behind thick concrete shields. Routine and maintenance operations should be performed remotely, as it is the case for the high active facilities of existing reprocessing plants such as the vitrification facilities of the La Hague plant. Auxiliary units needed for the maintenance (handling crane, special cells, etc.) will have to be implemented also in hot cells.

Technical equipment has to be designed to take into account this requirement, but the lack of knowledge concerning the sol-gel process and the absence of reference facilities for the whole process make difficult any detailed design study of the fabrication plant. It's the reason why this study deals only with the fabrication process and flow sheet.

Figure 1. The moderated target assembly



blue circle: moderator pin
red circle: MAs pin
green circle: spacer wire

Concentration unit

The concentration of the MAs nitrate solution coming from the partitioning unit of the reprocessing plant is equal to 0.45 g of americium and curium per litre, in this study. [5] Before feeding the head of the sol-gel conversion unit, the solution must be concentrated up to 360 g/l (1.5 mol MAs/l). Because of the high curium heat decay, such a concentrated solution releases up to 100 W/l, which is 10 times higher than the fission products concentrate of the La Hague plant and for which no industrial experience is available today. Any intermediate storage requires continuous cooling which is difficult to manage especially for incidental or accidental situations (e.g. partial loss of coolant flow).

A buffer storage of some weeks is deemed necessary between reprocessing plant and target fabrication in order to ensure sufficient availability of the whole process. Storage of diluted solution would require very large tanks, and therefore a two steps concentration process with intermediate buffer storage has been selected.

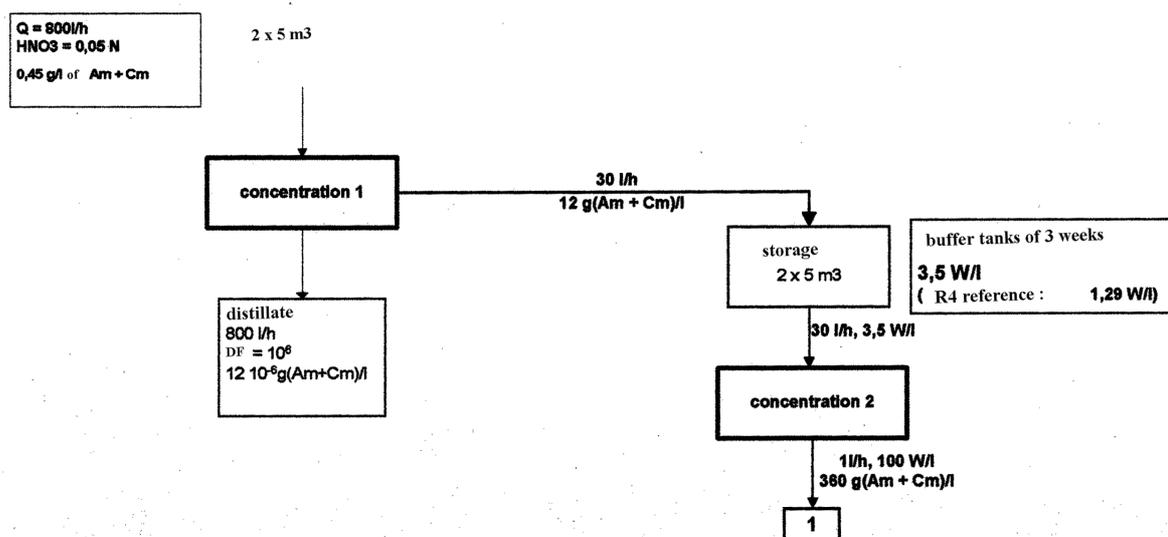
Figure 2 illustrates the concentration unit. Before the first concentration stage, the diluted solution is received in two buffer tanks of 5 m³ which alternatively feed the evaporator. The concentrated solution (12 g/l) releases 3.5 W/l and can be stored in two tanks of 5 m³, providing about 3 weeks of buffer storage which is considered as reasonable.

A second evaporator enables to concentrate the solution up to 360 g/l. The highly concentrated solution feeds directly the conversion step.

Conversion unit

The conversion step is the most difficult one to design, because direct synthesis of spherical particles of mixed americium and curium oxide is required in order to avoid further steps of powder mixing and granulation. These latter steps generate contaminant, fine and disperse dust, make the process complex by adding equipment for mixing, pressing and grinding and are thought to be very penalising when americium and especially curium have to be handled.

Figure 2. Concentration unit



Among the different known conversion processes (oxalic, ammonia, carbonate, peroxide, thermal denitration) the sol-gel process (ammonia-based conversion) appears to be the most suitable for the synthesis of mixed oxide microspheres or granulates. Other co-precipitation or thermal denitration processes leads generally to fine powders (particle size less than 10 μm).

Different variants of sol-gel processes have been used in the past in the nuclear field. They all consist in hydrolysing droplets of actinides nitrate solution with ammonia, but they differ in the way of gelation. The internal gelation process is certainly the most commonly used for the fabrication of microspheres of oxide, nitride or carbide fuels. Droplets of actinides nitrate solution are internally hydrolysed by the mean of chemical agents (in general, HexaMethyleneTetraAmine or HMTA) introduced in the solution and thermally decomposed to produce ammonia. The thermal decomposition can be induced in a hot silicon bath, where the droplets fall down or in a micro-wave furnace. For the external gelation, droplets of actinide solution fall down in the ammonia bath to be hydrolysed by diffusion of ammonia from the surface to the centre of the droplet. Although the external gelation process has been used successfully in the past for the fabrication of Am-pellets (SUPERFACT experiment, [6]) the internal gelation has been selected because this process should allow to limit the liquid waste volume, especially when microwave furnace is used for gelation.

The sol-gel process has been developed for the synthesis of uranium, thorium or plutonium based compounds but has never been used for the synthesis of particles of americium and curium compounds. The transposition is not obvious for three main reasons:

- hydrolysis of americium and curium (oxidation state III in solution) is not as easy as that of thorium, uranium or plutonium (oxidation state IV or VI);
- chemical agents used in the process, like HMTA or urea, could be decomposed by radiolysis or decay heat of curium. To avoid a premature degradation of organic compounds during the mixing of chemical reagents and before the droplets formation, an important adaptation of the process should be necessary;
- recycling of liquid waste, produced in significant quantities (additives, cleaning solution, solvents, etc.), is an additional problem to be solved.

Figure 3 gives a possible and schematic view of the conversion process, taking into account the flow rates given in the previous paragraph. Some comments are added concerning the nature of liquid and solid waste and the development studies required to validate the flow sheet. Figure 4 gives the industrial flow sheet of the process.

The actinide solution is cooled before dispersion to avoid a premature gelation. The mixed solution is dispersed through a vibrating nozzle to form droplets and through the microwave furnace to initiate the gelation. Spheres fall down into a cleaning bath of diluted ammonia or water to eliminate any traces of decomposition products of gelation agents (formaldehyde), ammonium nitrates, HMTA and urea in excess. Spheres are then dried in a column of hot (180°C) and humid air and calcined in a furnace at 800°C. If necessary for the further mixing step with magnesia, they are sintered to increase the particle robustness.

The MAs oxide coming from the conversion unit is received in a cooled hopper to be transferred to the pellets and pins fabrication unit.

Pellets and pins fabrication unit

Pellets fabrication is based on the conventional MOX fuel fabrication process, but is entirely set up in hot cells, remotely operated. The problem associated with curium decay heat have to be properly addressed. Figure 5 gives a scheme of the process.

The first step of the pellets fabrication consists in proportioning MgO and (Am, Cm)O₂ spheres to meet the composition requirements. Magnesia is poured out into a hopper and transferred into the powder blend station. (Am, Cm)O₂ spheres are transferred from the conversion unit as well. Both constituents are metered and introduced in the mixer and mixed till good homogeneity is obtained.

The mixed powder feeds through a hopper the mechanical press. The organic lubricant, needed to make easier and reproducible the pellet pressing, is not mixed to the powders before the pressing as usually performed in the MOX fabrication, but coats automatically the press dies. That is to avoid the risk of lubricant radiolysis and green pellets degradation during intermediate storage. Green pellets are stored in a molybdenum sintering boat. Some pellets are automatically extracted from the batch for visual and dimensional inspection and returned to the boat.

Green pellets are sintered in a continuous furnace. After sintering, pellets are controlled, sorted and characterised (chemical analysis, ceramographs, etc.). Pellets batches meeting the specifications are stored in plates in order to prepare the pin loading.

Pellets are laid out in columns of controlled length on a rack and placed in front of the fixed pin. A jack pushes the pellets column into the pin. The plenum spring is then introduced and the upper plug is welded under helium atmosphere. Welding is controlled, pins are cleaned and transferred to an intermediate storage in racks.

Figure 3. Conversion process of Am+Cm solution

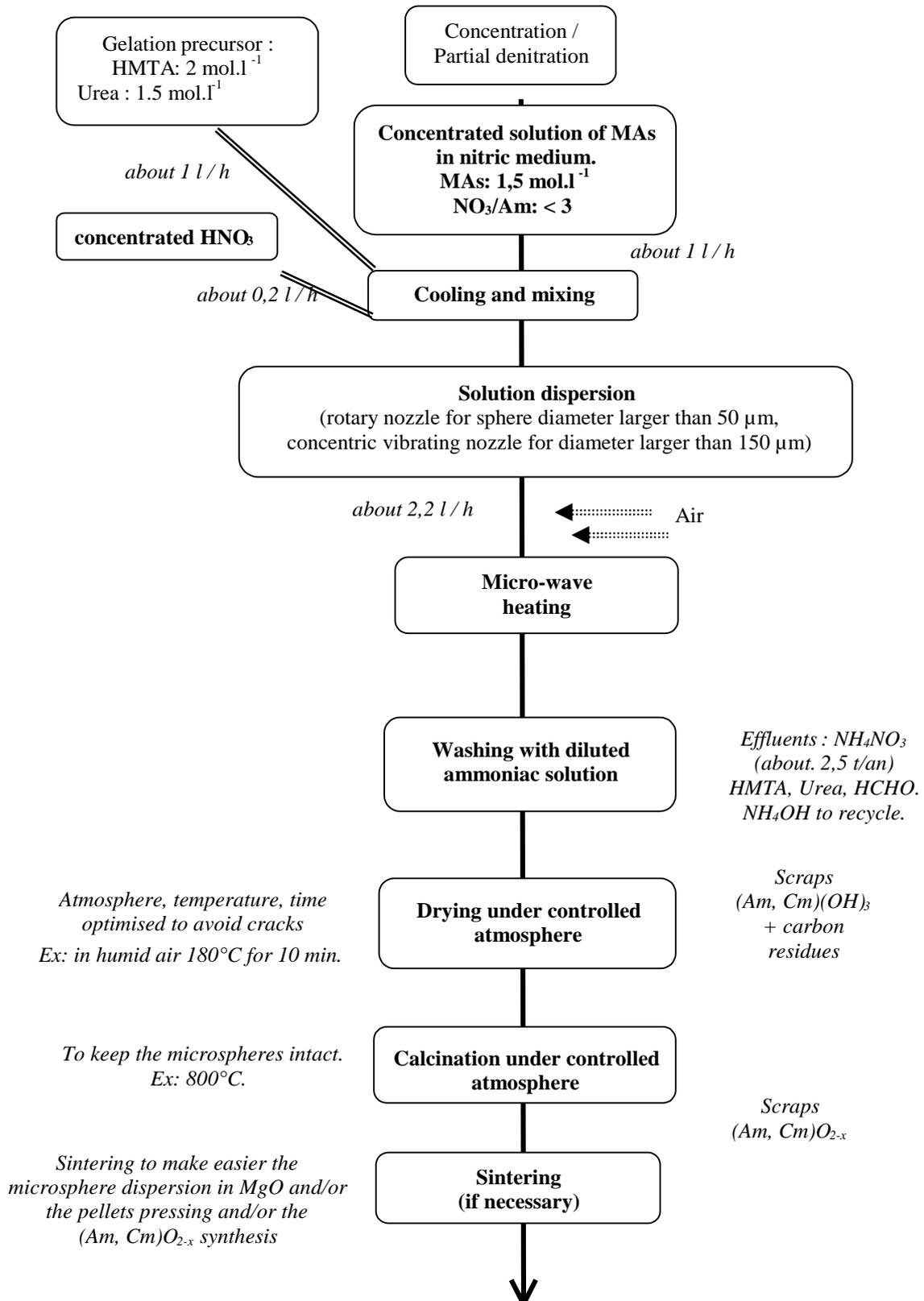
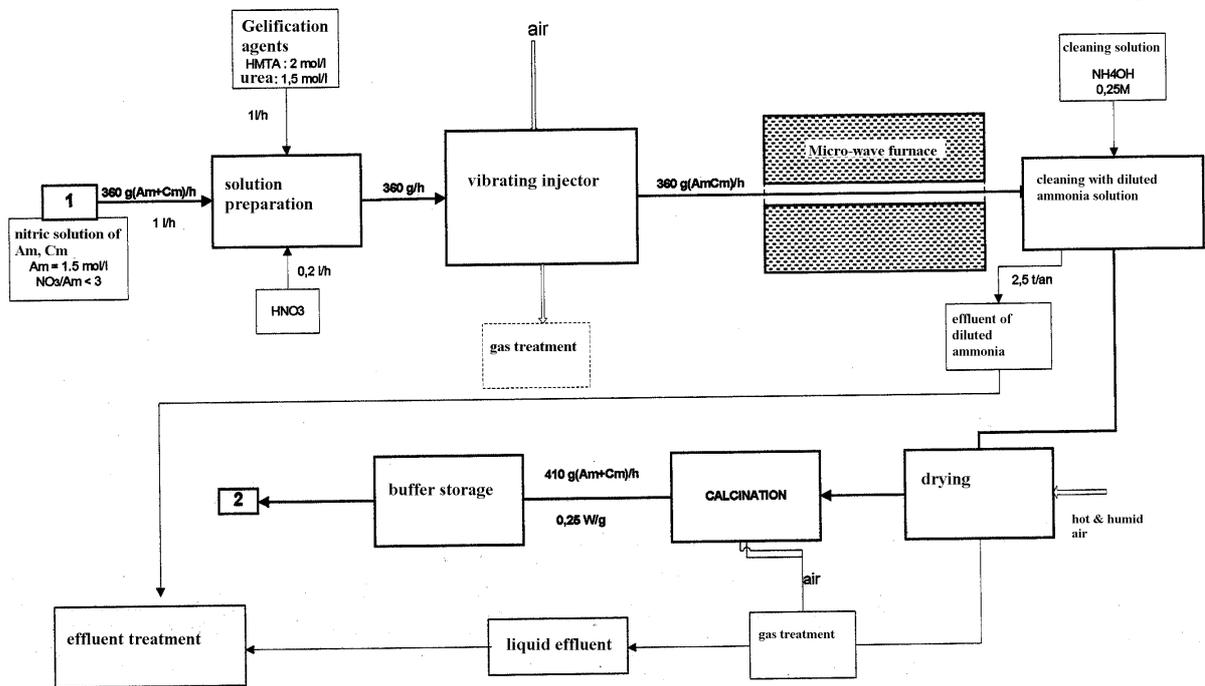


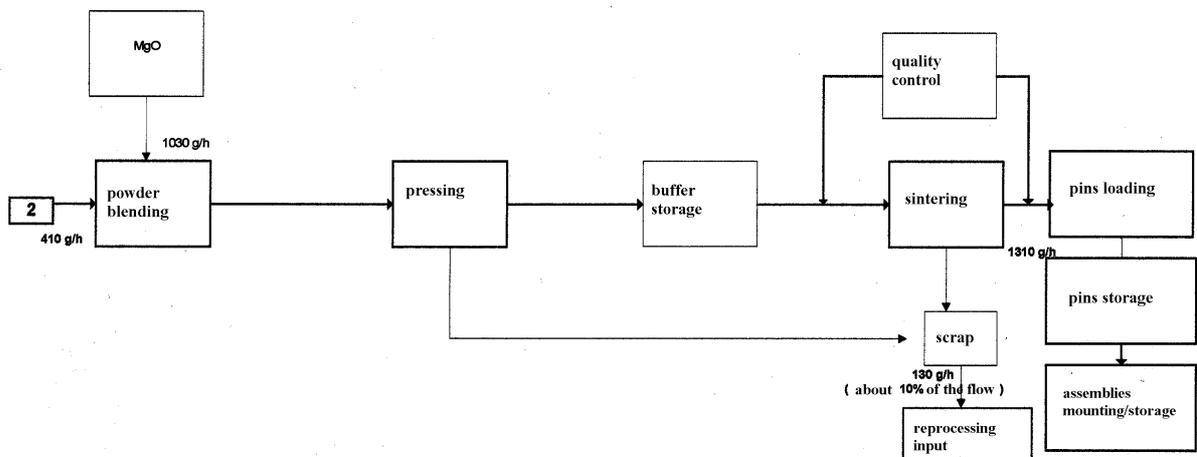
Figure 4. Conversion unit



Assembly mounting unit

Pins racks and the other structural materials (wrapper tube, moderator pins) are gathered in the assembly mounting unit. Pins are unloaded one by one to form the bundle. The bundle is attached to the assembly bottom and hung up to be introduced in the wrapper tube.

Figure 5. Pellets and pins fabrication



The upper part of the assembly is then fixed on the wrapper tube, welded and controlled. The neutron activity forces into manufacturing the assembly in shielded remote facility, what is penalising on looking at the fuel element size (the EFR assembly overall length is 4 800 mm). A continuous cooling is needed as well to keep the structural materials at a suitable temperature.

Waste treatment and scraps recycling

The sol-gel conversion process generates significant quantities of effluent or products of radiolysis and thermal decomposition, as mentioned on Figure 3. There is no experience or information concerning the treatment of such waste so that an extensive R&D programme would be required to minimise and manage them. Concerning the actinides hydroxides or oxides scraps, a recycling at the head of the reprocessing or conversion processes should be possible, since these facilities are located on the same site.

The powders mixing should not generate any reject other than retention losses.

Discarded green or sintered pellets could be recycled in the reprocessing plant head-end to be dissolved and reprocessed. The recycling in the pellets fabrication unit by grinding and mixing with magnesia and MAs oxide is not possible if macro-dispersion of (Am, Cm)O₂ is requested.

Pins loading and welding and assembly mounting should normally not produce waste since mechanical solutions are available to repair.

Conclusions

The technical feasibility of a fabrication facility of americium and curium targets is still far from the demonstration stage. Heat decay and neutron activity of curium lead to great difficulty of implementation of any industrial process. Tanks, pipes, fabrication tools, pins, assemblies, etc. will have to be continuously cooled, intermediate storage to be avoided or at least limited and the whole process should be implemented under heavy concrete shielding and remotely operated.

The sol-gel conversion process stands out as the most promising one because of the need of fabricating MAs oxide microspheres to obtain pellets with advanced macro-dispersed microstructure. However it is clear that this choice is speculative since it is not demonstrated that the sol-gel process is compatible with the decay heat and the neutron emission of curium.

The development of simple, compact and robust fabrication processes appears to be a challenge. However it is clear that simplification and compaction of fabrication processes unavoidably leads to make difficult advances on target microstructure: complex microstructure, like macro-dispersed structure, involves in general additional fabrication and control steps. That means that the expected benefit of advanced microstructure with regard to the target behaviour under irradiation must be weight against the technological impact on the fabrication process.

An important R&D effort is therefore necessary to investigate innovative processes. VIPAC or SPHEREPAC processes, where pins are directly loaded with the granulated powder from the conversion step, appear to be attractive candidates. However, development of co-conversion processes to synthesise particles of mixed MAs oxide or better still, particles of mixed inert matrix and MAs oxide is necessary.

CEA, in collaboration with RIAR and ITU, has been carrying out studies on VIPAC MgO + AmO₂ targets from pyrochemical processes since September 2001. Main objectives of the 2-years programme is to investigate 1) the feasibility of synthesising AmO₂ by precipitation from molten chlorides and separating americium from rare-earth elements, 2) the process to fabricate VIPAC MgO+AmO₂ pins and 3) the behaviour under irradiation of VIPAC target pins. In case of promising results an irradiation programme in BOR60 will be planned for demonstration.

Similar R&D activities, but based on wet processes and focused on co-conversion processes of MAs nitrate solution into microspheres or granulates of mixed (Am, Cm)O₂ oxides and inert matrix, should be planned in Europe as well. A R&D project will be proposed by CEA and European partners in the frame of the 6th FWP.

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