FEASIBILITY OF THE FABRICATION OF AMERICIUM TARGETS

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Abstract

The paper compares the processes used at ITU for the fabrication of americium targets for transmutation: powder mixing process, sol-gel method and the infiltration by an active solution of inactive pellets. The advantages of the latter processes, related mainly to the lower level of dust formation, are stressed. Moreover, the radiological constraints on the fabrication as a function of Am content and of selected fabrication process are evaluated. As conclusion, the feasibility of Am target fabrication has been demonstrated on a laboratory scale, based on experimental results evaluation. The penalties due to radiological constraints in a semi-industrial process are acceptable. The future developments consist in the construction of a laboratory fully dedicated to minor actinides fuel pins or targets fabrication.

Introduction

In the frame of the P&T programme, the transmutation of separated minor actinides and fission products requires the necessary step of manufacturing targets to be irradiated in specific reactors.

Fabrication of Pu-bearing fuels (MOX) both for LWR's or FBR's has now been mastered industrially, in Belgium, France, Germany, and Great Britain using powder blending processes. However, when fabrication of much more radioactive materials, like americium, curium or long-lived fission products is considered, dust production and consequent radiation exposure to personnel could limit the applicability of these mechanical processes. Therefore, alternative fabrication routes have been envisaged. The final selection of the fabrication routes relies on following evaluation parameters:

- Their technical feasibility.
- The quality and quantity of wastes produced.
- The accumulation of the dose to the personnel during fabrication and handling of the materials.
- The irradiation behaviour of the fuels and targets produced.
- The economical feasibility

In this paper, the first three aspects will be dealt with.

For the fabrication of fuels or targets containing highly radioactive elements, the following alternative processes can be compared with the mechanical blending of powders.

Gelation techniques

Gelation techniques have been developed and are still in use, in particular in Switzerland (PSI), Germany (ITU), and Russia.

These methods involve the dissolution of the starting materials (alternatively, if located at the reprocessing plant, the active solutions are readily available) followed by controlled droplet to particle conversion for the production of flowable microspheres. These are then pressed into pellets. The process produces little dust and can be automated readily.

These methods are particularly well suited in the frame of P&T strategies because of the high radio-toxicity of the active materials, but also of the limited capacity required in the manufacturing chains.

Hybrid gelation-blending methods

With hybrid methods we consider the combination of gelation techniques and mechanical mixture, for example particles made by gelation are mixed with inert matrices in the form of powders. These hybrid methods present similar advantages to the pure gelation methods, and allow larger output capacities.

Novel techniques

Advanced techniques are being developed. One of them is called INRAM (Infiltration of Radioactive Materials) [1,2]. It has been used for other applications in non-nuclear ceramics and for isolation of radioactive wastes, and has recently been applied for Am target fabrication. This process involves the dissolution of the active material and its infiltration into a porous (non-active) medium. Infiltration into porous green pellets drastically reduces the number of production steps involving handling of highly radioactive materials.

Americium Target Fabrication at ITU

General Requirements and Specifications

The term "Americium targets for transmutation" does not define the product to be manufactured, even in general manner. Today, it just means that cylindrical pellets containing a (variable) amount of americium diluted in a preferably non-uranium matrix have to be manufactured and loaded in fuel pins, in geometries of either LWR's or FBR's! There are no well defined specifications for the pellet, but well a series of criteria (neutronic and materials properties, solubility, reaction with coolant …) that should lead to the selection of the dilution materials.

It is not the aim of this paper to review all alternatives of supporting matrices examined in the world. Let us just state that ITU, in the field of transmutation research, concentrates its efforts on following materials:

- Oxides of Am (stabilised with Zr), diluted in inert matrices of the types spinel ($MgAl_2O_4$), MgO, or stabilised ZrO_2. The type of microstructure (homogeneous mixture, or so-called macro-masses of Am-rich particles in the matrix) is a parameter that must be considered in the development stage.
- A mixed oxide of AmO_{2-x} and ThO_2 chosen per analogy with UO_2, which presents the advantage of an easier licensability of the product.
- Nitrides, for their good thermal conductivity. However, this subject, based on specific processes, will not be discussed in this paper.

The elaboration of specifications of such new materials is made following continuous exchanges between the designers, the manufacturers, and the final users, namely today the operators of the experimental facilities like PHENIX or Material Testing Reactors.

In case of Am targets, a particular difficulty exists in the quality control of the fuel, because of the specific nature of the compounds.

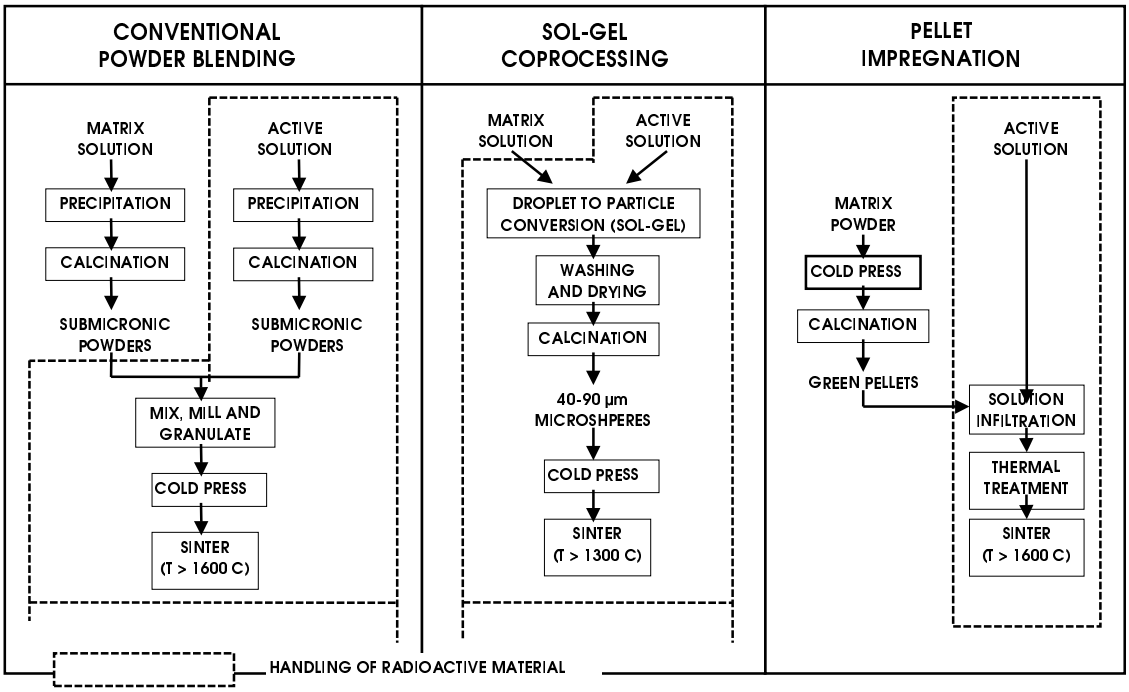
Sol-gel method

The sol-gel coprocessing process (Fig. 1), developed originally at ITU for the production of MOX fuels, has also been used to fabricate U-Pu-Am-Np oxide fuels for the transmutation and incineration of americium and neptunium. In this procedure, a droplet to particle conversion process is used to produce beads (ϕ = 20 - 300 μm) with high mechanical stability, so that dust formation is eliminated. Furthermore, the free flowing beads reduce the fabrication steps and facilitate automation of the process.

At the heart of the sol-gel beads production step is an ammonia precipitation of the metal hydroxides from an aqueous solution, which requests the need of the active solution preparation. Waste solutions can be recovered and recycled.

Today, the sol-gel method is used for the development work on the fabrication of $(ZrAm)O_2$ + inert matrices, using cerium as a simulant for Am. The fabrication parameters have to be adopted to obtain good quality pellets (dimensions, stability, density, visual aspect ...), and taking into account the wishes of the designer to have the Am particles distributed heterogeneously in the matrix (to the contrary of all present requests for industrial MOX fabrication!). This is therefore a particular challenge.

Figure 1. **Pellet fabrication procedures**



INRAM method

The flow sheet for the fabrication of INRAM pellets is shown and compared with the sol-gel and mechanical blending methods in Figure 1. The main advantage of this method is that matrices with low or zero activity can be fabricated and formed into the required shape in an unshielded facility. The matrix material is then introduced into the shielded glove-box and immersed in a solution of the infiltrant in a controlled way. Thereafter the resulting material is treated thermally to convert the infiltrant into the desired chemical form, and sintered to produce the product pellets. As no precipitation or washing steps are required within the shielded area, the radioactive wastes produced in the process are negligible. The fabrication process can also be performed in facilities combining remote handling for the fabrication steps and standard glove-box technology should intervention be required.

The infiltration process relies on the action of capillary forces to draw the solution into the pores of the host material. The application of this process to the fabrication of transmutation and incineration targets requires that the pellet is insoluble in the solution containing the infiltrant and that the infiltrant should be easily convertible into its desired chemical form.

The concentration of the second phase introduced into the pellet can be controlled by adjusting the concentration of the infiltrant solution to the porosity of the pellet. Immersion of green spinel pellets with 50% porosity in a 400 g/L Am solution yielded a final product with 11 $^{w}/_{o}$ Am. Tailored distributions of the infiltrant can be obtained by complete immersion of the pellet and control of the immersion time and/or by sealing selected surfaces of the pellet.

The infiltration of radioactive materials (INRAM) method was used to produce two fuel pins, containing 11 $^{w}/_{o}$ Am in spinel for the EFTTRA-T4 [3] irradiation in HFR Petten, which is now completed. In the pre-fabrication phase, test pellets with 9 $^{w}/_{o}$ Am were fabricated. A relatively uniform Am distribution throughout the pellets was obtained for the pre-fabrication test pellets. The pellets for the EFTTRA-T4 irradiation were produced in exactly the same way, but the total Am content was 11 $^{w}/_{o}$. The Am distribution within the pellet was not as uniform as in the pre-fabrication tests. In particular, α-autoradiography showed a roughly cylindrical symmetric shell (200 μm thick) within the pellet in which the Am content was higher than the surrounding regions. EPMA revealed an Am concentration of 14 $^{w}/_{o}$ within this band and 9 $^{w}/_{o}$ elsewhere. This is most probably due to a diffusion process during the thermal treatment in the steps involving liquid to solid Am nitrate and/or nitrate to oxide conversion. The guaranteed homogeneity requires further investigations of these effects. Optical microscopy of the samples has shown that using the INRAM method, the Am containing particles have a diameter of 2 - 3 μm.

Technical feasibility of the fabrication

Handling of highly radioactive materials and the fabrication of targets containing minor actinides should be considered at both laboratory and industrial scales. At the ITU a minor actinide laboratory is being developed to cover the needs of the ITU and its partners for the fabrication of fuels and targets in the framework of test irradiation programmes (e.g. EFTTRA and CAPRA) and to make tests for future industrial applications. The handling facilities (see Fig. 2) consist of shielded cells combining the protection of a hot cell with the flexibility of a glove-box. These hybrid cells are equipped with manipulators and biological protection (water and lead) for direct handling of minor actinide containing fuels and targets. When the minor actinides are removed from the box, or shielded *in situ*, access to the boxes can be obtained using conventional glove-box procedures. In this way, equipment can be exchanged or modified within the cell or even the entire glove-box behind the protection wall can be replaced. Thus the high level of flexibility necessary for research and development can be maintained. The laboratory is currently being modified and cells incorporating powder preparation (sol-gel and INRAM methods), pellet pressing, sintering, control and pin fabrication are being built.

Fabrication methods based on sol-gel or infiltration (INRAM) could provide alternative means for the preparation of targets for transmutation and incineration. Due to the low levels of dust formation, both processes are compatible with the handling facilities in use at the ITU. Further investigations are required to provide process data for these procedures which would permit their scaling up for industrial production. A possible flow sheet for an industrial scale application of the INRAM process is shown in Fig. 3, which shows a rough evaluation of the flow of materials required for each step, corresponding to the quantity of americium which would be available annually from 50 LWR's [3] in a strategy that would produce about 1 ton Am/year. In this flow sheet, assumptions have been made regarding the Am content in the fuel (10 $^{w}/_{o}$), fuel length and equipment capacities. The number of different equipment required is given on the flow sheet. Only one immersion tank would be required. Industrial application will require extensive if not complete automation and

adequate shielding, and further evaluation assessments of the radiation dose. A similar evaluation could be done for a plant relying on sol-gel method.

Figure 2. **Minor actinide laboratory: lay-out**

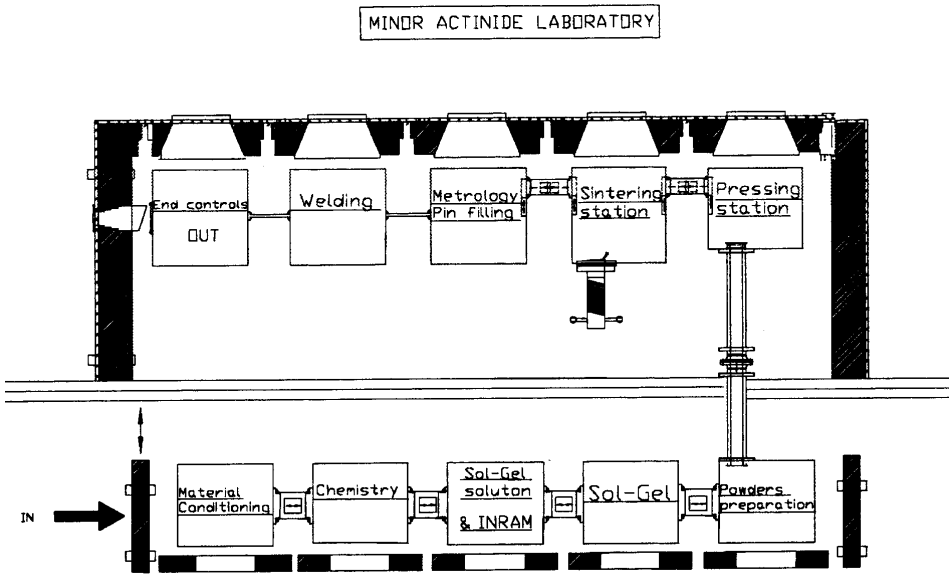
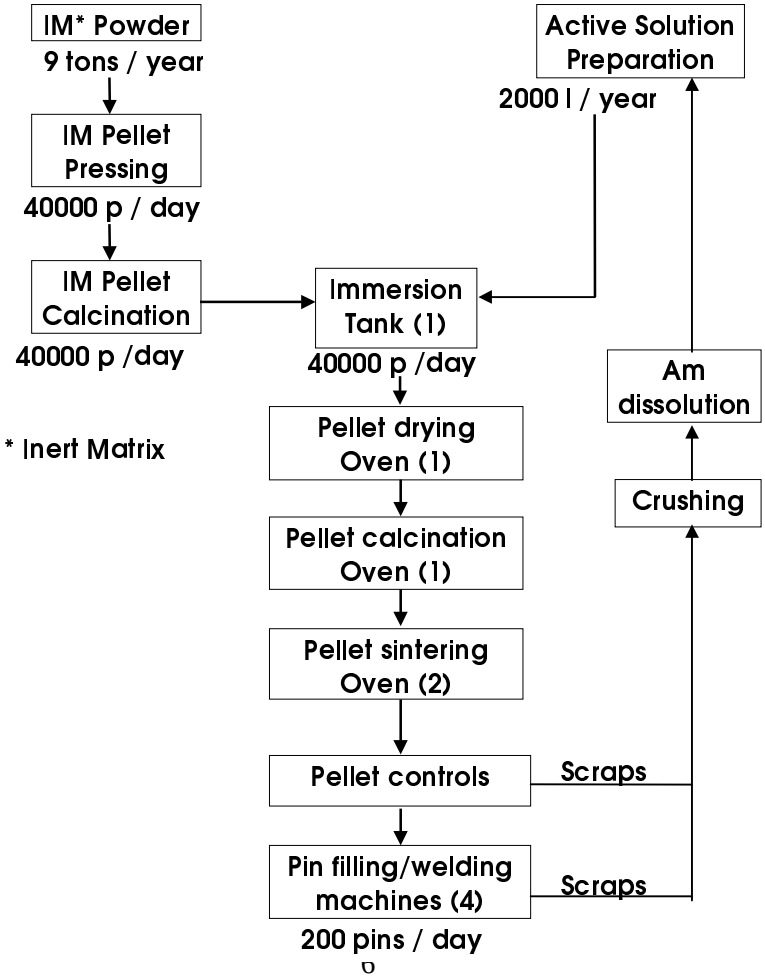


Figure 3. **Flow sheet for americium target fabrication INRAM process 1 ton Am/year**



Radiological constraints

Due to the high activity of the minor actinide materials, the feasibility of the fabrication of Am targets depends very much on the additional radiation protection measures necessary for each process step as well as for the storage of the primary Am material and the transport of the fabricated product.

The semi-industrial process implies, in comparison with the laboratory techniques:

- The handling of bigger quantities of materials which induces higher dose rates and the need of increased automation of the process.
- The adoption of maintenance procedures due to production constraints.
- Additional costs associated with the radioprotection and automation equipment.

The radiological constraints are a function of the content of americium in the targets and of the density of the support material.

Comparison with standard MOX fuel

Specific conditions of fabrication and transport of Am target pins for recycling in LWR cores are analysed by comparison with the current (U-Pu) MOX fuel fabrication at BELGONUCLEAIRE - Dessel (Belgium). The equivalent dose rates are evaluated by means of 3D calculation techniques validated on benchmarks and experimental results obtained in the fabrication chain and around a transport container as well as from some measurements performed on a SUPERFACT pellet [6].

The front-end stages of the fabrication process are the most critical ones because the heat and radiation sources are concentrated in the primary material. One example is the comparison of the storage of the same amount of pure PuO_2 and AmO_2 powders in a shielded cavity of a typical MOX fabrication plant, which shows an increase of the total dose rate in front of the doors by a factor 82, for the americium oxide case.

Further in the process, the handling of a target pin, designed for insertion in a PWR fuel assembly with 20% AmO_2 in an inert matrix for instance, induces a total dose rate behind a 10 mm thick Plexiglas layer, which is about 1500 times higher than when handling a PWR MOX pin with 7.8% Pu. Indeed, the dose due to PWR MOX is weak, but for any case of AmO_2 content higher than about 2% in such a target pin, a reinforced protection made of lead glass (or steel) is necessary to fulfil the current dose rate limit criteria at some distance from the glove box. The reduction of the autoabsorption of gamma rays in the inert matrix compared to an UO_2 matrix plays an important role.

At the back-end stage, the transport of a large number of target pins, either towards the fuel assembly mounting hall or already grouped in a Am-dedicated assembly, would require a big reinforcement of the lead shielding of the cask licensed for fresh MOX fuel, so that it could be more appropriate to use a cask designed for irradiated fuel, but it would be oversized and very heavy.

Feasibility evaluation

The comparison with the MOX fuel fabrication conditions applies to the Am-target pin fabrication by the conventional powder blending process. The conclusion is that it is preferable to establish a fabrication chain dedicated to Am targets in shielded cells with some automation equipment, using specific transport containers, instead of adapting standard MOX fuel equipment.

The required additional protections are quite feasible, although they are cumbersome and they increase the costs. However, the penalties due to these constraints in a semi-industrial process seem quite acceptable.

As the radioprotection constraints are mostly conditioned by the radioactive sources, proportional to the Am content of the pellets, and by the density of the pellet matrix, the fabrication technique has only a very limited influence on the order of magnitude of the dose rate due to a target rod during handling and transport. Consequently, the additional protections for those operations are similar for each technique.

But the fabrication technique has an important effect on the secondary radiations due to dust, waste, and possible contamination. The advantages of the INRAM technique are namely due to the reduced number of fabrication steps involving highly radioactive materials and to the liquid nature of the infiltrant, so that the hazard of radioactive dusts can be minimised, as well as the risk of contamination within the glove boxes.

Conclusion

For the fabrication of fuels and targets containing Minor Actinides (in particular Am), processes that produce minimal quantities of dust and waste are recommended.

In particular, ITU has demonstrated on a laboratory scale, the feasibility of the infiltration (INRAM) technique consisting of the infiltration of the active solution (Am nitrate solution) into a non-active matrix (a porous pellet in this case) by capillary forces. Tests are still required to permanently avoid the heterogeneity sometimes observed in the americium distribution, which were probably due to diffusion during thermal treatment.

Theoretical studies performed during the last years, as well as experimental results on ion-bombardment of inert matrices, however, predict that for Am-containing targets macro-dispersions of the fissile phase are to be preferred to micro-dispersions, in order to limit radiation damage in the matrix due to alpha-decay and stopping of fission products [3].

The current research at ITU in this field is as follows:

- improvement of INRAM methods: to apply this method on porous beads instead of pellets to obtain higher actinide contents (up to 40 $^{w}/_{o}$) and guaranteed homogeneous distributions
- macro-dispersion fuels: to fabricate inert matrix fuels containing Am in inclusions of sizes > 50 μm by mixing Am containing spheres (produced by gelation methods) with inert matrix powders.

Both processes are the reference for the minor actinides fabrication laboratory currently under construction.

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