Minor Actinide Laboratory at JRC-ITU: Fuel fabrication facility

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Abstract – *The Minor Actinide Laboratory (MA-lab) of ITU is a unique facility for the fabrication of fuels and targets containing minor actinides. It is of key importance for research on Partitioning and Transmutation (P&T) in Europe, as it is one of the only dedicated facilities for the fabrication of MA containing materials, either for property measurements or for the production of test pins for irradiation experiments. In this paper a detailed description of the MA-Lab facility and the fabrication processes developed to fabricate fuels and samples containing high content of minor actinides is given. In addition, experience gained and improvements are also outlined.*

INTRODUCTION

The world-wide interest in management options for the back-end of the nuclear fuel cycle has lead to many new ideas about the treatment of the actinides that are present in spent nuclear fuel. Partitioning and Transmutation (P&T) and Partitioning and Conditioning (P&C) are presently considered to be promising techniques. P&T is a method to reduce the radiotoxic inventory of the nuclear waste to be disposed of in a geological repository. It essentially means that the actinides are separated from the spent fuel by aqueous or pyrochemical methods and re-irradiated in a nuclear device (a critical or subcritical reactor) to transform them into short-lived or stable isotopes by fission. Different scenarios are investigated in which the actinides are incorporated in fuel or in inert matrix targets as individual elements or as a mixture. As little experience exists on such fuels or targets, extensive research is required to establish fabrication methods, to measure the basic properties and to test the irradiation behaviour. P&C is a method to reduce the radiological effects of geological disposal of the nuclear waste. Again the actinides (and fission products) are separated from the spent fuel, but in this case they are incorporated in a material with an improved chemical durability for specific geological conditions. Similar to P&T, fabrication methods must be developed, basic properties of the material must be measured and the durability must be tested.

Due to the nature of the tests, the research programmes for P&T and P&C need to be performed with significant quantities of materials. This leads, however, to a significant increase of the radiation protection measures because the handling of gram quantities of minor actinides such as americium and curium requires special shielded facilities to protect the workers from the irradiation. As shown in Table I, the most relevant minor actinide nuclides have a relatively high specific activity and upon decay they emit not only alpha radiation but also gamma radiation. In addition, some of the nuclides emit neutrons originating from the spontaneous fission. Alpha radiation has a very low penetration depth and pure alpha emitters can therefore be handled in glove boxes in which radiation is shielded by plexiglas windows and the rubber of the gloves

Nuclide	Specific Activity (Bq/g)	Decay characteristics				Spontaneous fission
		Alpha		Gamma		
		Energy (MeV)	Probability	Energy (keV)	Probability	
^{237}Np	2.610 10^{7}	4.79	0.48	29.4	0.15	
^{241}Am	1.271 10^{11}	5.49	0.852	59.5	0.359	●
^{242m}Am	3.598 10^{11}	5.20	0.004	49.4	0.002	
^{243}Am	7.391 10^{9}	5.28	0.880	74.7	0.674	●
^{243}Cm	1.911 10^{12}	5.79	0.735	277.6	0.140	
^{244}Cm	2.997 10^{12}	5.80	0.770	42.8	0.0002	●

These air-tight glove boxes also serve as containment for the activity. The penetration depth of gamma and neutron radiation is much larger. Therefore special protection is required: heavy materials such lead, steel or concrete for gamma radiation and light materials such as water or polyethylene for neutron radiation.

GENERAL LAYOUT OF THE MA-LAB

The MA-lab consists of seven glove-boxes with protection walls forming two separate chains. A schematic lay-out of the Ma-Lab is shown in Figure 1. Its protection wall to shield the operators from the gamma and neutron radiation has 50 cm water and 5 cm lead as protection at working level and 10 cm polyethylene and 2 mm lead above. Based on the thickness of the water and lead wall, the mass limits have been calculated for a number of radionuclides to be handled in the MA-lab (see Table I). This calculation is based on the assumption that 98% of the particles of the powder have an AMAD greater than 10 µm. However, for most of these nuclides the operating licence of ITU dictates the limiting mass, with the exception of ^{231}Pa (photons) and ^{244}Cm (neutrons).

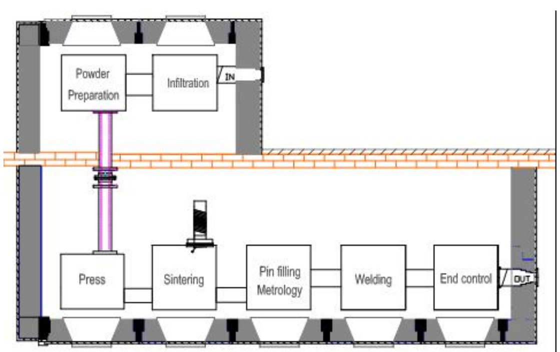

Fig. 1. General lay-out of the MA laboratory

Telemanipulators are used for normal operation. In addition, extensive automation with remote control has been included in the design. However, the cells are in fact standard glove boxes that permit manual intervention in the absence of radioactive sources, in case of maintenance and repair. The fabrication in the MA-lab is based on advanced liquid-to-solid processes, described in detail below, to avoid dust-formation and its accumulation in the cells, which would make manual intervention very difficult.

TABLE I. Limiting masses

Isotope	Limiting mass (g)	Criterion
^{231}Pa	10	Shielding
^{237}Np	9600	License
^{241}Am	2000	License
^{242m}Am	0.1	Shielding
^{243}Am	34	License
^{243}Cm	0.14	Licence
^{244}Cm	5	Shielding

DESCRIPTION OF THE FABRICATION PROCESS

As a result of the shielding, process simplification and automation are necessary. In addition, the waste generated should be minimized. The fabrication procedure should not generate dust, which could collect on the surfaces of the glove boxes and the equipment therein, so that operator intervention is facilitated and radiation exposure to the personnel minimized. For these reasons, an advanced dust-free fabrication process developed at ITU has become the reference fabrication process in the MA-lab. This process is a combination of the gel supported precipitation [1,2] and the porous bead infiltration techniques [3]. This process not only meets the criteria listed above but also it is flexible and easily adapted to the requirements and specifications of new fuel and target compositions.

Powder Preparation and Treatment

A flowsheet of the fabrication process is shown in Figure 2. The actinide material is entered into the chain as powder where it is dissolved in nitric acid to the required metal concentration. Porous beads are produced outside the MA-lab, either in a cold laboratory if the beads are made of inactive materials or in an active chain, if the beads contain Pu. Then they are introduced into the Ma-Lab where they are infiltrated with the nitrate solution of the required minor actinide. The solution is added dropwise until the incipient wetness point is reached. The infiltrated beads are thermally treated in a calcination furnace. After this process step, the infiltration can be repeated multiple times if higher actinide content in the beads is required. Optionally the beads are mixed with a second phase [4] , the matrix, in a specific mixing device. Once calcined the powder (mixture) is compacted into pellets which are sintered in controlled atmosphere in a sintering furnace. Thereafter, the pellets are inspected visually using a camera, weighed and their dimensions are measured in an optical device to determine their density. The best pellets are selected and positioned on a loading tray for pin filling. For specific properties measurements samples in the form o disks are also produced.

Pin Filling and Welding

Together with the necessary components (e.g. inactive pellets, spring) the pellets are loaded into the fuel pin, and following welding of the end cap X-ray images of the weld and the pin are made. After the surface contamination of the pin is measured the pin is removed from the chain.

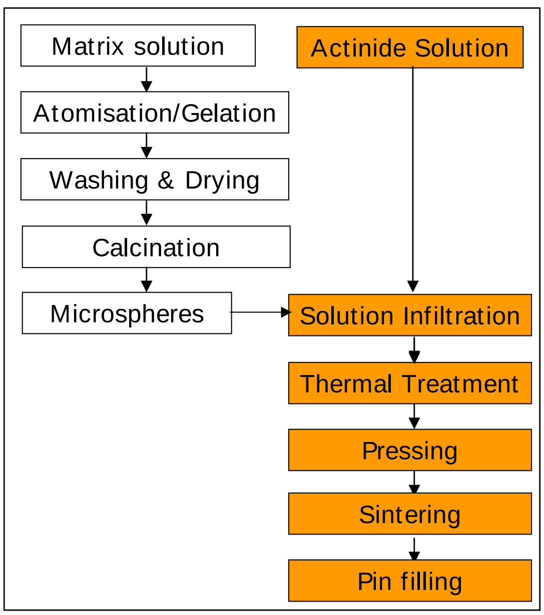

Fig. 2. Schematic flow sheet of the reference fabrication process of the MA-chain.

IRRADIATION EXPERIMENTS

ITU is actively involved in several irradiation programs both in the High Flux Reactor (HFR) at Petten and the Phénix reactor in Marcoule. Therefore, fuels and pins for irradiation have been fabricated and fully characterized. In addition, post-irradiation experiments will be performed in the ITU Hot Cells laboratories. Table II summarizes the main characteristics of the fuels fabricated in the MA-Lab for the irradiation experiments.

TABLE II. General Characteristics of the Fuels

HELIOS 2	$(Am_{0.06},Zr_{0.78},Y_{0.16})O_x$ Homogeneous
HELIOS 3	$(Am_{0.20}, Pu, Zr_{0.66},Y_{0.14})O_x$ Homogeneous
HELIOS 4	$(Am_{0.20}, Zr_{0.66},Y_{0.14})O_x$ + 72%vol Mo Macrodispersion - 90-130µm
HELIOS 5	$(Pu_{0.8}Am_{0.2})O_2$ + 84%vol Mo Microdisperdion - 20-120µm
FUTURIX 5	$(Pu_{0.8}Am_{0.2})O_2$ + 86%vol Mo Microdisperdion - 20-120µm
FUTURIX 6	$(Pu_{0.23}Am_{0.25}Zr_{0.52})O_2$ + 60 % vol Mo Microdisperdion - 20-120µm

Irradiation experiment in Phénix - FUTURIX

The purpose of the FUTURIX-FTA international irradiation program is to demonstrate the feasibility of burning minor actinides in dedicated reactors including critical electricity-generating fast reactor transmuters or hybrid systems (ADS). Eight pins including metallic, nitride, CERMET and CERCER forms prepared by the ANL, LANL, ITU and CEA, respectively, are being irradiated in the PHENIX reactor since May 2007. This experiment will provide essential data concerning behaviour under irradiation and will allow qualification and validation of models developed to predict fuel performance. At ITU, two molybdenum based cermet fuels have been fabricated for this experiment.

Irradiation experiment in HFR- HELIOS

The main objective of the HELIOS irradiation test is to study the in-pile behaviour of U-free fuels in order to gain knowledge on the role of the microstructure and of the operating temperature on the gas release and on fuel swelling. The test matrix contains both homogeneous, zirconia-based ceramic compounds and heterogeneous compounds, based on MgO and molybdenum as inert matrices. The HELIOS irradiation experiment is planned to begin in 2008 in the HFR and will last for 300 EFFD.

CONCLUSIONS

The MA-Lab is a unique facility for the fabrication of fuels and targets containing minor actinides. Since it became active in 2004 several fabrication campaigns have been successfully completed. The flexibility of the fabrication process permits the production of homogenous or composite fuels.

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