MA Laboratory

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1. Introduction

The safe disposal of highly active waste and spent nuclear fuel dominates the nuclear debate in several countries. The major issue is the potential risk due to the long-term radiotoxicity of the Transuranium elements. Whereas Uranium and Plutonium recovery and re-use are technically feasible and is practised in a number of countries, a major challenge continues to be the minor actinides, such as Neptunium, Americium and Curium. These long-lived radiotoxic elements should preferentially be removed (partitioned) and destroyed (transmuted) before disposing nuclear waste in a final repository.

The main purpose of ITU´s activity in that field is to define and characterise the most suitable materials for transmutation of Transuranium elements, in order to assess the technical feasibility of P&T. This objective requires further investigation on the manufacturing of fuels and targets for irradiation testing, and their material property determinations. For that purpose the Minor actinide Laboratory has been constructed in the Nuclear Fuels Unit at ITU. It allows the fabrication and control of fuel pellets and experimental fuel pins containing large amounts of highly radioactive minor actinides, for testing in irradiation experiments.

The P&T research at ITU is performed in close relationship with our European partners, and further research integration, including non-European countries, is made within the ongoing 5th framework Programme [1].

2. P&T strategies and need for experimental equipment

To investigate the effects of different P&T strategies on the radiotoxicity reduction, three cases have been considered in a study made at ITU [2], in comparison to the open cycle. The resulting radiotoxicity curves (based on ICRP72 reference, and for 50 GWd/tHM burnup) are shown in Figure 2, in which the cross over point indicates the time at which the radiotoxicity of the waste reaches the reference level. The following results have been obtained:

- The open cycle: the spent fuel is directly sent to the long-term storage with no P&T. It takes therefore 130,000 years before the radiotoxicity reaches the reference level.
- The full multi-recycling of Pu as well as Am and Cm with high overall efficiency of P&T processes (99.5% for Pu and 99% for Am + Cm). The cross over point is 500 years. If the Cm is left in the waste, this time is extended to 1000 years.
- The full multi-recycling of Pu as well as Am and Cm with less overall efficiency of P&T processes (99.5% for Pu and 95% for Am + Cm). The cross over point is 1000 years.
- The partial multi-recycling: multi-recycling of the Pu (99.5% of P&T efficiency), and one single recycling of the Am and Cm. In this case the Am and Cm are transmuted in once-through targets in a fast reactor, and for them 90% of P&T overall efficiency is foreseen. Thus the cross over point is around 1,500 years. In this strategy, we can also consider leaving the Cm in the waste, and then 3,000 years are required.

Based on these results it can be concluded that P&T of Pu and Minor Actinides can help to reduce the time during which nuclear waste should be isolated from the biosphere from 130,000 years to between 500 years and 1,500 years. The fission products radiotoxicity curve gives the theoretical limit to the total radiotoxicity reduction in the case that all the actinides are partitioned and transmuted i.e. no losses. This time is about 270 years.

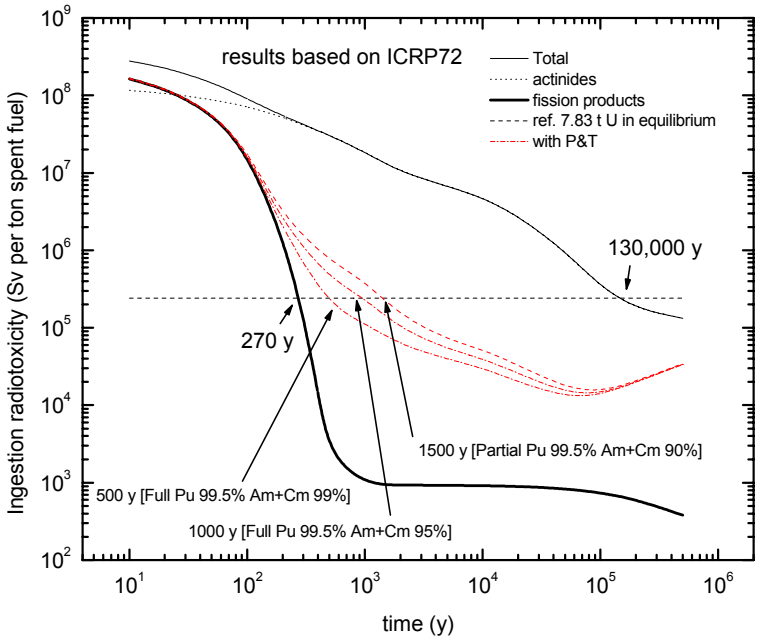

Figure 1: Ingestion Radiotoxicity of one ton used nuclear fuel based on ICRP72 effective dose coefficients [2]

It should be noted that a timescale of up to 10,000 years is consistent with the licensing of underground repository of nuclear waste such as Yucca Mountain [3], and, for the different P&T strategies, the period during which the waste are still radiotoxic (above the reference level) are included in this timescale.

These strategies require the knowledge and demonstration of advanced aqueous reprocessing of spent oxide fuel, to recover Pu, Np, Am and Cm. The reprocessing in current (but modified) installations requires high separation rates for Pu and Np (99.9%), but lower ones for Am and Cm (99 %). This seems feasible from present research results using the DIAMEX process [4]. One open question is the treatment of the Cm. If the Cm is not transmuted while the Am is, then the separation of the Cm from Am has to be done, which needs further experimental demonstration. In this case Cm must be conditioned and decay to Pu, which could be further transmuted if necessary.

After separation, Pu and Np are recycled as MOX fuel, whereas Am and perhaps Cm have to be conditioned preferably in stable U-free targets for their irradiation in fast neutron systems for their transmutation by fission. The development of the facilities and fabrication processes are going on mainly at CEA and ITU [5,6] for Europe, and the feasibility of their fabrication, transmutation and good irradiation behaviour has been partially demonstrated [7]. New experiments planned in Phenix and HFR will further increase the present data base [8] and more experiments will still be required before considering these new compounds for licensing in commercial installation and for large scale utilisation. This can be achieved within about 15 years taking into account the long lead-time needed in reactor materials developments. A particular technical question to be resolved in the partial multi-recycle strategy is the high transmutation rate required: the amount of actinides still present in the irradiated targets should be less than 10% of the actinides initially loaded. This requires high burning rates, and the demonstration that the materials, fuel and cladding, can sustain these limits without failure. To prepare the fuels for these new experiments, adequate laboratories are required, such as the Minor Actinide Laboratory, which is now in the commissioning status.

3. The Minor Actinides Laboratory

The Minor Actinide Laboratory (MA-lab) of the Institute for Transuranium Elements is a unique facility for the fabrication of fuels and targets containing minor actinides such as americium and curium. 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. It can also be used to prepare ceramic materials for long-term disposal in case partitioning of some long-lived elements would be followed by conditioning (P&C strategy).

The MA-lab consists of ten glove-boxes with protection walls forming two separate chains. The main chain consists of 7 glove-boxes. Its protection wall to shield the operators from the gamma and neutron radiation emitted by the isotopes of these elements, have 50 cm water and 5 cm lead as protection at working level and 10 cm polyethylene and 2 mm lead above. The second chain, consists of 3 glove-boxes behind a protection wall of 10 cm polyethylene and 5 mm lead. The limiting masses are:

- 34 grams ^{243}Am (shielding),
- 5 grams ^{244}Cm (shielding),
- 10 grams of ^{231}Pa (shielding),
- 150 grams of ^{241}Am (license).

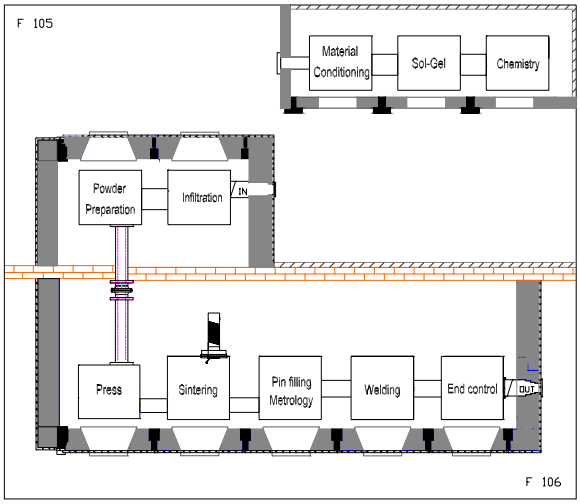

Fig. 2: Schematic drawing of the MA-lab layout.

Fig. 3: Operation by telemanipulators

Telemanipulators are used for normal operation. In addition, extensive automation with remote control and robots has been included in the design. However, the glove boxes 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 to avoid dust-formation and its accumulation in the cells, which would make manual intervention very difficult. The infiltration process (INRAM) is the reference process for the MA-lab. In this process, which has been developed for nuclear applications at ITU, the minor actinides are infiltrated into porous particles, which can be pressed directly into pellets, or infiltrated directly into porous pellets [6]. The production free-flowing (thus dust-free) powders by SOL-GEL processing for direct compaction to pellets is also possible in the chain, but will be limited to the use of Am, to avoid Cm-contaminated liquid wastes.

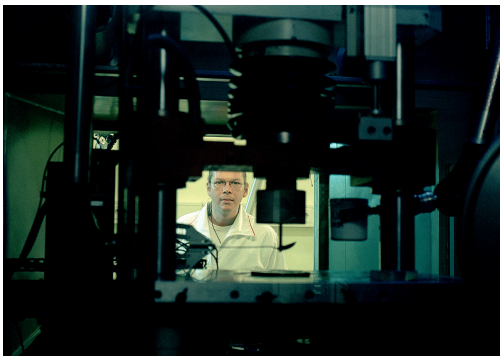

Fig 4: The MA-lab press in Box 6.

The facilities in the MA-lab permit the fabrication of fuel pins with a maximum length of 1 metre. State-of-the-art TIG welding is installed and the fuel pins can be examined with high resolution X-ray radiography. The pin filling, welding and control operations (dimensions, leak tightness, X-ray) are remotely achieved as well.

The construction of the MA-lab has been funded by the Joint Research Centre of the European Commission. The design and construction has been has been made by ITU staff, with assistance of local and international companies. In the coming years its will be used for the realisation of international projects within the Framework Programmes of the European Commission and for the fabrication of fuel pins for international clients. The programmes foreseen are:

- in 2003: Fabrication of 3 $(Am,Zr)O_2$+MgO pins for an irradiation programme in PHENIX (contract with CEA, CAMIX-COCHIX);
- in 2003 - 2004: Fabrication of $(Am,Pu,Zr)O_2$ and $(Am,Pu,Th)O_2$ and (Am,Zr)N pellets for materials characterisation (FUTURE and CONFIRM FP5 projects);
- in 2004: Fabrication of 3 (Am, Pu, Zr)O2 pins for HFR irradiation
- in 2004 - 2005: Fabrication of 1 (Am,Pu,U)O2 and 1 U-free CERMET pellets batches and welding of max. 8 pins for PHENIX (collaboration with CEA and DOE, proposal for FP6).

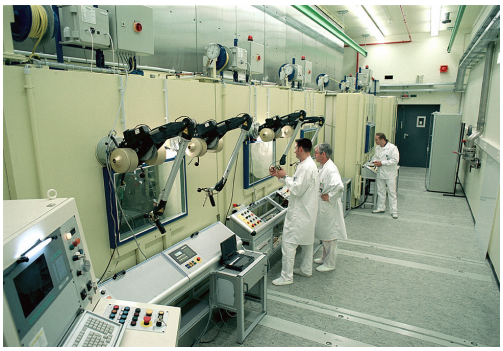

Fig. 5: The front of the cells 6-10.

4. Conclusion

P&T can achieve very large reduction factors in the radiotoxicity of radioactive high level wastes. If Pu and the minor actinides are extracted and re-irradiated for transmutation, the wastes can meet the reference radiotoxicity level after 500 to 3,000 years (instead of 130,00 years without P&T), depending on the strategy selected. However, extensive experimental programmes and large scale demonstration will be required before any commercial P&T can be implemented. Therefore, dedicated facilities are needed. They are mainly the transmutation reactors (experimental or prototype Fast Reactors or ADS, like MYRRHA), but also the fuel cycle facilities for advanced reprocessing and for the fuels and targets fabrication. The new MA Laboratory will serve this goal.

References

1. "Nuclear Waste Partitioning and Transmutation in the Euratom Fifth Framework Program" M.Hugon, AccApp/ADTTA, ANS Topical Meeting, Reno, NV, USA, Nov. 11-15, 2001
2. "Impact Limits of Partitioning and Transmutation Scenarios on Nuclear Waste Isolation Times", J. Magill, V. Berthou, J. Galy, R. Schenkel, H.-W. Wiese, G. Heusener, J. Tommasi, G. Youinou, JRC-ITU-NT-2002/24.
3. "Yucca Mountain Science and Engineering Report" US DOE, February 2002
4. "Advanced Reprocessing Research at ITU" J-P. Glatz, B. Christiansen, R. Malmbeck, M.Ougier, T. Inoue, T. Koyama, K. Kinoshita, GLOBAL 2001, 9-13 September 2001, Paris, France.
5. "Status of the European Program on ADS Fuels" S.Pillon, AccApp/ADTTA, ANS Topical Meeting, Reno, NV, USA, Nov. 11-15, 2001
6. "Transmutation of actinides: Qualification of an advanced fabrication process based on the infiltration of actinide solutions" A. Fernández, D. Haas, R.J.M. Konings, J. Somers, J. Am. Ceram. Soc. 85 (2002) 694
7. "10 YEARS EFTTRA: 1992-2001" D. Warin, R. Conrad D. Haas, G. Heusener, P. Martin, R.J.M. Konings, R.P.C. Schram, G. Vambenepe, GLOBAL'01, 9-13 September 2001, Paris, France.
8. "Fabrication of transmutation fuels and targets: the ECRIX and CAMIX-COCHIX experience " Y. Croixmarie, E. Abonneau, F. Desmouliere, L. Donnet, A. Fernández and R.J.M. Konings. Presented at the ERMT-II Seminar, Karlsruhe 26-27 September 2002 (to be published in J. Nucl. Mat.).