EFTTRA IRRADIATION EXPERIMENTS FOR THE DEVELOPMENT OF FUELS AND TARGETS FOR TRANSMUTATION

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ABSTRACT

The EFTTRA collaboration (Experimental Feasibility of Targets for Transmutation) between CEA (France), ECN (The Netherlands), EDF (France), FZK (Germany), IAM and ITU (European Commission) was launched in 1992, with the aim of performing joint experiments for the study of materials for the transmutation. Irradiations have started in parallel in the Phénix fast reactor in France, and in the high flux thermal reactor HFR in the Netherlands. One of these experiments, concerning technetium and iodine, has been completed ; post-irradiation examinations of the Tc metallic samples are performed by ECN, CEA and ITU, and a summary of the last results is presented. The other on-going EFTTRA experiments are described, with a report on the application of fabrication methods for matrices with up to 10% americium content. Finally, some considerations on the strategies for americium are given.

INTRODUCTION

In the field of the research on nuclear wastes management, the problem of the long-lived radioactive nuclides requires careful consideration. The possibility of separating and transmuting these long-lived radioactive nuclides, with the aim of reducing the radiotoxicity of the final waste, has to be investigated. To carry out such experimental or theoretical investigations in the frame of an international collaboration presents many advantages, one of them being to split the costs of the studies. Therefore, the decision to form the EFTTRA (Experimental Feasibility of Targets for Transmutation) group, devoted to technical problems, together with European partners, was taken [1].

EFTTRA PROGRAMME AND RESULTS

The EFTTRA programme presently consists of the study of the transmutation of Tc-99 (metal), and of the development of materials (inert matrices) for the transmutation of americium. One experiment on Tc (EFTTRA-T1) concerned also iodine.

Technetium

Metallic Tc rods have been produced and welded in 6 cylindrical target capsules (3 for HFR and 3 for Phénix), at ITU, for irradiation in HFR and in Phénix. The irradiation of 3 target capsules in HFR, an experiment called EFTTRA-T1, was completed in 1994, and "the first results of the post-irradiation examination at ECN, which have been presented earlier [2-4], show that the burn-up is about 6.4 % and that no swelling of the rods has occurred. Part of these irradiated samples have been sent to ITU and CEA for comparative examination ; the results from the three laboratories will be discussed and interpreted in common, and a detailed report will be published. The observations at ITU and CEA confirm the results obtained earlier at ECN that the irradiation had no significant effect on the structure of the material, and no swelling was detected. It will be checked if this good behaviour of technetium metal remains for higher burnup (see below, EFTTRA-T2). The distribution of the Ru concentration along the radius of the sample, determined at ITU by EMPA (Electron MicroProbe Analysis), showed the same sharp decrease as for the ECN sample, from ~ 13 % at the surface, down to 8 % at 0.1 mm depth and 6 % at 1mm depth (diameter of the samples : 4,8 mm). The value obtained at the surface of the sample is slightly below the ECN value : this can be due to the very steep slope of the concentration curve near the surface, or to the orientation of the sample. No significant variation of the Ru concentration was found along the axis of the samples. The PIE observations are in excellent agreement with the results of detailed KENO Monte-Carlo calculations, as performed at ECN [5].

Two technetium rods have been re-packed at ECN for a re-irradiation in the HFR. This irradiation (EFTTRA-T2) was started in February 1996 and will last for about 500 full power days during which a burn-up of more than 20% will be achieved.

The irradiation of the 3 other Tc target capsules is planned in the fast reactor Phénix (EFTTRA-F2) ; depending on the schedule of the Phénix reactor, the irradiation should start in 1997. The samples will be placed in the radial blanket of the reactor, in a thermalized neutron flux ; the moderator is CaH_2. The experiment aims at a transmutation of 15%.

Iodine

The iodine capsules (containing natural I-127) of the EFTTRA-T1 irradiation were examined at ECN [4,6]. The capsules containing CeI_3 and NaI did not show any degradation and the burn-up was 5-6 %, the capsules containing PbI_2 were heavily corroded and leakage had occurred. Taking also into account the information from the fabrication of the targets, NaI seems to be the best of the three candidate materials. However, because the necessity of iodine transmutation is still under discussion and the implementation of the transmutation of iodine in the fuel cycle is very complicated (large inventories, long transmutation half lives, vented pins concepts to release Xe pressure), the EFTTRA cooperation will stop its efforts on this element.

Inert matrices

The heterogeneous recycling of Am supposes that targets for the transmutation, with a high percentage of americium (10 to 40%), are produced, irradiated, and reprocessed. The choice of a suited inert matrix depends on its thermodynamic, physico-chemical, and mechanical properties, and on its behaviour under irradiation or during cooling.

The preliminary studies are done in the laboratory, for a first selection of matrices [7,8]. Techniques have to be developed for the fabrication of the materials ; if reprocessing by the Purex process is envisaged, the solubility of the matrix in nitric acid, possibly after crushing of the material, is an important factor : MgO was found to be the most easily soluble. Once the matrix has been fabricated, a range of parameters has to be measured : the ideal matrix material should have high thermal conductivity and melting point temperature, low creep and swelling properties, low activation coefficient, low neutron absorption, and should not react with the cladding or the coolant. Radiation damage can also be studied out-of-pile, by ion implantation. Table 1 shows the main criteria and the present situation on the evaluation of candidate (oxide) matrices. A similar study is now underway for nitride matrices.

For the study of the behaviour under irradiation of the candidate matrices, several experiments are planned. UO_2 may be added to the samples, to simulate the presence of americium oxide. The EFTTRA-T2 experiment, already mentioned in the above section on Tc, also includes the irradiation in HFR of samples of Al_2O_3, YAG ($Y_3Al_5O_{12}$) and spinel ($MgAl_2O_4$). A shorter, parallel irradiation, with also a sample of CeO_2, started at the same time in HFR (EFTTRA-T2bis), and should give a quicker indication of the behaviour of the matrices in a high thermal neutron flux environment.

The start of the EFTTRA-T3 experiment, in which a selection of inert matrices is mixed with UO_2, 20% enriched in U-235, is planned for the end of November 1996. Table 2 gives a list of the samples which have been selected for this irradiation.

Details on the EFTTRA-F1 and EFTTRA-F1bis experiments, also called Matina 1 and Matina 1bis, in the Phénix fast reactor have been given earlier [8-10] ; changes in the programme may occur, depending on the operation planning of the Phénix reactor, and of possible new orientations in the strategy.

Americium targets

The same preliminary studies on fabrication, properties, and reprocessing, have to be done with the matrix material containing americium. This was started with the AmO - MgO system [11].

The irradiation in HFR of a sample of americium oxide embedded in a spinel matrix, the EFTTRA-T4 experiment, is now under preparation. This experiment is partly financed by the European Commission, through the shared cost actions of the Framework Programme 1994-1998 on Nuclear Fission Safety. The samples have been produced by ITU, using an impregnation method developed in the frame of a research programme of the Institute on innovative fabrication techniques for the fuels of tomorrow ; a publication is under preparation, which will give detailed information on the technique. Dose measurements have been performed on the samples, giving indications on the protective measures to be considered for the future developments of the technique ; it can already be stated that the impregnation technique elaborated at ITU presents two main advantages in this respect, compared to the classical powder mixing method : the low amount of wastes produced, and the possibility to remotely control the process, i.e. to produce the samples with a good radiological protection of the operator.

The samples, containing 10 wt% of Am-241, will be encapsulated in a stainless steel cladding. The irradiation is planned to start in September 1996 ; the duration will be about 400 full power days, with a fluence of the order of 4×10^{26} m^{-2}. According to the calculations [12,13], the actinide density in the sample could be reduced by 35 %.

UNDERLYING STRATEGIES

The efforts of the EFTTRA group are concentrated on the technological aspects of the transmutation, namely the development of targets for the transmutation of technetium and americium. However, the connection of our programme with the studies on strategies cannot be denied : it has to be consistent with the possible options for the future, and the final choices will be made taking also into account our results on the technical practicability of the proposed solutions. As such, EFTTRA is participating in the shared cost action of the European Commission on the Possible P&T Strategies, where it will contribute to the work package on the assessment of the feasibility of advanced fuel or target fabrication.

In most of the countries using nuclear energy, no strategy has been chosen yet for the long term, concerning the back-end of the fuel cycle, i.e. the fate of the plutonium and of the minor actinides. Different scenarios have been defined, which could be classified according to the recycling scheme, considering the available reactor park : the CEA studied 4 scenarios, PWRs only (recycling of Pu and minor actinides in MOX fuel), PWRs and recycling in fast reactors, PWRs with Pu monorecycling and subsequent recycling in fast reactors, or fast reactors only. Sometimes, separate dedicated devices for the transmutation are proposed, like for the Japanese "double-strata" system [14], based on fast reactors or hybrid (accelerator) systems. The scenarios include also the possible evolution of the nuclear energy in the coming years : decrease, stability, or increase (possibly with the development of new reactor technology). The final aim of the study of the various scenarios is an appreciation of their impact on the reduction of the radiotoxicity of the end waste disposal. For each scenario, many elements have to be investigated, like the consequences on core operation, on safety, on cycle operation ; the reactor parameters have to be optimised, and the materials tested. There are also other items entering in the appreciation of the value of a scenario :

- should it be a strategy for today, with the present technology, or a solution for the future ?
- weight of the economic factor

- capacity of reducing the losses from reprocessing
- is there a request for reprocessing, or is once-through permitted?
- technological possibility to go to high burn-up
- can the reprocessing technique be different from PUREX ?

Concerning the field of interest of the EFTTRA group, i.e. the transmutation of americium, Table 3 shows the spectrum of envisaged possibilities. The advantage of an inert matrix is the reduction of the production of plutonium isotopes ; but a UO_2 matrix is easier to reprocess, has the best behaviour under irradiation, and it has been shown that the extra radiotoxicity produced is negligible. If the once-through solution is adopted, there will be no reprocessing, but very long irradiation times will be necessary, with the problem of finding suitable materials.

CONCLUSION

Before a definitive choice of the right strategy for the future can be made, the study of the different scenarios must be pursued, in order to have enough elements of decision available. EFTTRA is participating in this effort, with a range of complementary irradiation experiments, both in a thermal and in a fast neutron spectrum, including the fabrication tests, the determination of the properties of the candidate materials, and the post-irradiation examination programme.

REFERENCES

[1] J.-F. Babelot, H. Gruppelaar, G. Mühling, C. Prunier, M. Rome, M. Salvatores, EFTTRA, A European collaboration for the development of fuels and targets for the transmutation, in Proceedings of the Third International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Cadarache, December 12-14, 1994, NEA/P&T Report N°13, 1995, pp. 110-115.

[2] J.-F. Babelot, R. Conrad, W.M.P. Franken, J. van Geel, H. Gruppelaar, G. Mühling, C. Prunier, M. Rome, M. Salvatores, Development of fuels for the transmutation in the frame of the EFTTRA European collaboration, in GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles, September 11-14, 1995, pp. 524-529.

[3] R.J.M. Konings, E.H.P. Cordfunke, G. Dassel, W.M.P. Franken, H. Gruppelaar, J.L. Kloosterman, Irradiation tests of transmutation of the fission products technetium and iodine, in GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles, September 11-14, 1995, pp.1631-1637.

[4] R.J.M. Konings, W.M.P. Franken, R. Conrad, J.-F. Gueugnon, J.-C. Spirlet, Transmutation of technetium and iodine- Irradiation tests in the frame of the EFTTRA cooperation, to be published in Nuclear Technology.

[5] J.L. Kloosterman, J.A. Hendriks, R.J.M. Konings, Transmutation of technetium in the Petten HFR : a comparison of measurement and calculation (to be presented at this meeting).

[6] R.J.M. Konings, Transmutation of iodine: results of the EFTTRA-T1 irradiation test, J. Nucl. Mat. (submitted).

[7] N. Cocuaud, T. Duverneix, R. Mazoyer, Y. Philipponneau, J.M. Adnet, J.P. Dancausse, Experimental studies on heterogeneous recycling, in GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles, September 11-14, 1995, pp. 530-537.

[8] C. Prunier, Y. Guérin, M. Salvatores, A. Zaetta, Concept and experimental studies on fuel and target for minor actinides and fission products transmutation, in Proceedings of the Third International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Cadarache, December 12-14, 1994, NEA/P&T Report N°13, 1995, pp. 224-228.

[9] C. Prunier, Y. Guérin, J. Faugère, N. Cocuaud, J.M. Adnet, The CEA SPIN programme : minor actinide fuel and target aspects, in GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles, September 11-14, 1995, pp. 506-515.

[10] N. Chauvin, J.-L. Faugère, C. Morin, J.-F. Babelot, In-pile research on minor actinide burning, in GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles, September 11-14, 1995, pp. 1691-1699.

[11] S. Casalta, K. Richter, C. Prunier, A study of AmO_2-MgO system for americium target transmutation in fast reactors, in GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles, September 11-14, 1995, pp. 1725-1731.

[12] J.L. Kloosterman, Incentives for transmutation of americium in thermal reactors, report ECN-R--94-022, October 1994.

[13] W.J.M. de Kruijf, Calculational study on irradiation of americium fuel samples in the Petten High Flux reactor, report ECN-R--94-027, February 1995.

[14] T. Mukaiyama, Importance of Double Strata Fuel Cycle for Minor Actinide Transmutation, in Proceedings of the Third International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Cadarache, December 12-14, 1994, NEA/P&T Report N°13, pp. 30-39.

Table 1 : Evaluation of candidate inert matrices for the transmutation of minor actinides

Candidate oxide matrices for M.A. transmutation in a PWR

Technical steps	MgO	$MgAl_2O_4$	Al_2O_3	CeO_2	Y_2O_3	$Y_3Al_5O_{12}$	$CePO_4$	$ZrSiO_4$
Manufacturing	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Basic properties	Yes	Yes	Yes	Yes	Yes	Yes ?	Yes	No
Neutron activation	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
H_2O compatibility	No	Yes	Yes	Yes	No	Yes	?	?
In pile behaviour	Yes	Yes ?	No	?	?	Yes ?	?	?
PWR compatibility	**No**	**Yes ?**	**No**	**Yes ?**	**No**	**Yes ?**	**?**	**No**

Candidate oxide matrices for M.A. transmutation in a Fast Reactor

Technical steps	MgO	$MgAl_2O_4$	Al_2O3	CeO_2	Y_2O_3	$Y_3Al_5O_{12}$	$CePO_4$	$ZrSiO_4$
Manufacturing	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Basic properties	Yes	Yes	Yes	Yes	Yes	Yes ?	Yes	No
Neutron activation	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Na compatibility	Yes	Yes	No	No	Yes	Yes	No	No
In pile behaviour	Yes	Yes ?	No	?	?	Yes ?	?	?
FR compatibility	**Yes**	**Yes ?**	**No**	**No**	**Yes ?**	**Yes ?**	**No ?**	**No**

Table 2 : EFTTRA- T3, irradiation of candidate inert matrices in HFR Petten

level 1	$CeO_2 + UO_2$	CeO_2	$Y_2O_3 + UO_2$	Y_2O_3
	$CePO_4 + UO_2$	$CePO_4$	$Y_2Al_5O_{12} + UO_2$	$Y_2Al_5O_{12}$
level 2	$MgAl_2O_4 + UO_2$ (sol-gel)	$MgAl_2O_4$ (sol-gel)	$MgAl_2O_4 + UO_2$	$MgAl_2O_4$
	YN + UN	YN	ZrN + UN	ZrN

(shaded) also for CAPRA-programme

Table 3 : Possible strategies for the transmutation of americium

	thermal spectrum		fast spectrum	
	once-through	reprocessing	once-through	reprocessing
inert / heterogeneous	few % Am	> 20% Am	X	> 40% Am
UO_2 / heterogeneous	X	> 20% Am	X	> 40% Am
MOX / homogeneous	X	few % Am	X	few % Am