System-immanent Long-lived Radioisotope Transmutation

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Abstract

The Institute for Transuranium Elements of the Joint Research Center at Karlsruhe has been involved in the study of minor actinides for about twenty years. Facilities exist to prepare and analyse the material properties of MA - containing targets prior to or after irradiation.

The past studies related to MA partitioning and transmutation were aimed at recycling MA's in the presently developed nuclear power stations: LWR and/or FR. Long-lived radionuclides such as 99Tc and 129I need higher neutron fluxes than those available in nuclear power stations to achieve short transmutation times.

The ongoing research is focused on the need for a broader knowledge of basic thermodynamic and chemophysical data primarily for the design of MA -, 1- and Tc - containing targets and to evaluate newly proposed extractants for partitioning under "hot" conditions.

Introduction

In the European Institute for Transuranium Elements, advanced fuels for fast reactors have been developed and tested over the last 25 years. In the 70's, minor actinide (Np, Am) containing mixed oxide fuels were designed and successfully irradiated in fast reactors: KNK II and Phenix [1, 2]. The composition of the fuel covered the homogeneous as well as the heterogeneous recycle of minor actinides (see table 1). The advantage of such fuels would have been an increased energy generation of about 4 % which would compensate for the extra fuel make-up costs, increase the breeding ratio and prolong burn-up reactivity [3,10]. A recent study showed that the changes in the void and Doppler effect are acceptable from the safety point of view [4].

Tab. 1: Irradiation Experiments with Different Minor Actinide Fuels

Fuel type	Reactor	Status
$(U_{.73}Pu_{.25}Np_{.02})O_{1.97}$ $(U.73PU.25241Am.02)\ O_{1.97}$ NpO_2 $(^{241}Am_{.5}U_{.5})O_{1.92}$	KNK II	irradiation completed
$(U_{.77}Pu_{.21}Np_{.02})O_{1.97}$ $(U_{.77}Pu_{.21}{}^{241}Am_{.02})O_{1.97}$ $(Np_{.45}U_{.55})O_{2.00}$ $(Np.2241Am.2\ U_{.6})O_{1.95}$	PHENIX	irradiation completed

When this work was initiated in the seventies the economic aspect of minor actinides was more important than the recently discussed ecological advantages of partitioning and transmutation schemes. Since then the introduction of fast breeder reactors has been postponed for at least 40 years. Under these changed circumstances we felt it necessary to reexamine all options of the transmutation of long-lived radionuclides. Hence a workshop was organized by the Institute in October 1989 [5] to assess the technical feasibility of the proposed transmutation procedures. A review of this workshop has been given [6].

System-immanent Recycling of Long-lived Radionuclides

For a convincing ecological transmutation scheme other radionuclides as well as the minor actinides have to be considered. Not only the easily migrating monovalent cation of pentavalent neptunium (NpO_2^+) (and hence its parent nuclide, ^{241}Am) have to be considered but also other monovalent ions of the fission products, the most prominent of which are the anions of ^{129}I (I^-) and 99Tc (TcO_4^-).

So far the recycling of plutonium in the so-called self-generated mode has been introduced for LWR [7]. The other nuclides mentioned above show a very different behaviour when recycled in the thermal neutron spectrum of a PWR (see table 2). The transmutation half-lives of ^{237}Np and ^{241}Am are acceptable with 4.3 and 0.9 years respectively. However the underlying process is neutron capture leading finally to ^{238}Pu and ^{242}Pu which when recycled together with the self- generated plutonium would worsen its handling properties due to enhanced neutron radiation by the spontaneous fission of ^{238}Pu. On the other hand it would denature the plutonium helping to prevent a misuse of the fissile material. The table refers to an unloaded spent fuel of a PWR fuel initially enriched with 3.5 % ^{235}U. In the case of self-generated Pu recycling, the amount ^{243}Am will be enhanced by a factor of 25 [7]. The recycling of this nuclide produces mainly ^{244}Cm. When ^{244}Cm is recycled in a thermal neutron spectrum, which is known as a strong neutron emitter, ^{252}Cf builds up significantly, thus making the handling of the spent fuel almost impossible. For this reason ^{244}Cm should not be fed back to LWR power stations but rather be burnt in the harder neutron spectra of fast reactors.

In order to transmute the two fission products 99Tc and ^{129}I in a reasonable time period a thermal neutron flux of about 10^{15} n/cm^{-2} see-l would be needed. Reactors of this type have only been developed for research purposes, but they would be the best means of transmuting these nuclides. Fast reactors though, having a higher neutron flux, are not efficient because of the correspondingly lower neutron capture cross-sections of the fission products.

Instead of recycling Np and Am in a LWR they could be recycled together with Cm in fast reactors. This second option of system-immanent minor actinide recycling would be valid for a wide deployment of fast reactors and correspond to our earlier concepts of advanced fast reactor fuels [81.

Tab. 2: Long-living Radiotoxic Nuclides in PWR

(33 GW_{th} d/t, initial ^{235}U = 3.2 w/o, 3 x 10^{13} n $s^{-1}cm^{-2}$)						
	^{99}Tc	^{129}I	^{237}Np	^{241}Am	^{243}Am	^{244}Cm
Discharge Concentration [g/t]	780	170	440	67	84	24
Halflives [y]						
Tota l	12	35	4.3	0.88	9.2	13.1
Capture	12	35	4.3	8.7/0.98	9.2	53
Fission			>104	230	>104	610
Decay	>105	>107	$>10^6$	433	>103	18

Research and Development for the System-immanent Recycling of Minor Actinides

There exists some know-how on how to recycle Np in light-water reactors since this nuclide was a source of ^{238}Pu used in pacemakers. The recycling of Am in light- water reactors is difficult because of the high resonance shielding of ^{242}Am. Earlier irradiation tests show that the burn-up in the outer region of the fuel is so strong that the material embrittles [9]. Therefore new concepts must be developed which would e.g. consist of annular fuels. The development of Am - and Np - containing fast reactor fuels has been mentioned above. The two fuel compositions cover the extreme cases of self-generated recycling with 2 % minor actinides and the heterogeneous concept with up to 50 % of minor actinides. Concentrations in between have to be tested, since they exhibit additional advantages for reactor operation [4, 10, 11]. Cm - containing fast reactor fuels have not so far been tested. If the concentration of this element is kept in the permille range no major changes in the irradiation behaviour of the fuel is expected. However the handling properties of such a material will require heavier shielding during fuel make-up.

Apart from the need to develop high-thermal-flux reactors the corresponding targets for the transmutation of Tc and I have to be designed. Such reactors would transmutate Np and Am 10 times faster than LWR'S and therefore the recycle of MA's in such reactors must be considered too. It should be recalled that the concentrations of Np and Am are of the same order asTc and 1 (see table 2). In a high neutron flux the fission of ^{238}Np will contribute significantly to the transmutation process.

in order to design the needed new fuels required for thermal reactors and/or improve those for fast reactors, more basic data of the thermal physical behaviour and material properties are needed. So far our studies have concentrated on the phase diagrams of americium oxides [12], neptunium oxides [13] and on a limited sector of the quaternary oxides of Am - Np - U -0 relevant to the fuels described above. To assess the compatibility of mixed oxides with sodium the Gibb's free energy was determined [14] and the thermal conductivity over a temperature range from 600 OC to 2000 °C measured [15]. This information, together with known data, were sufficient to predict the irradiation behaviour of minor actinide fuels. For more comprehensive future studies however, a broader data base will be needed. Such studies (see table 3) are envisaged at present and have to some extent been started.

Tab. 3: MA - related Research

Basic Research ● Integral neutron cross-sections ● Thermodynamic data ● Chemophysical data
Applied Research ● Simulation experiments ● Characterisation of targets ● Fabrication of targets ● Irradiation experiments ● Partitioning studies

There is a common problem for all transmutation options concerning the partitioning of minor actinides from high level waste. The various processes are only capable of separating the lanthanide and actinide groups together [16]. Recently a new derivative of phosphine oxide (TRPO) was compared in its extraction properties to the chemical of the TRUEX process: CMPO [17]. The synthesis of other compounds however may lead to more specific separations. In the USSR a process based on Carbolyle has been developed, which achieves the lanthanide / actinide separation in a single extraction process [18]. A comparative "hot" test of the newly proposed partitioning scheme should be made on a lab scale as outlined under [17] for TRPO.

Conclusion

The recycling of long-lived radionuclides in existing nuclear power stations and the development of a high-thermal-flux reactor offer the possibility of transmuting such nuclides into short-lived or stable elements. When compared to more advanced transmutation procedures such as minor actinide burner [19] or neutron spallation sources [20, 21], one finds that system-immanent recycling would require less research and development effort. The needs for partitioning are unchanged.

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