RADIATION DOSE LEVELS IN THE HANDLING OF MINOR ACTINIDE FUELS

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ABSTRACT

An experimental and theoretical assessment is presented of the radiation dose levels encountered in the fabrication and discharge from fast reactor of minor actinide-containing fuels (SUPERFACT fuels). The gamma radiation dominates the total dose.

INTRODUCTION

The management of nuclear waste, either in the form of discharged spent fuel or high level waste (HLW) from fuel reprocessing, is confronted with the radiotoxicity potential of transuranium elements and fission products. The long-term radiotoxicity is dominated by ^{241}Am, ^{240}Pu and ^{237}Np, and a partitioning of these nuclides from the waste would bring the anthropogenic radiation dose close to the uranium ore mined to produce the nuclear fuel (1). Therefore, the possibility of partitioning these minor actinides (MA) out of the waste and transmuting them into hazardless stable nuclides has been proposed and its technical feasibility is being studied (2,3).

The inclusion of MA, and particularly americium, in a fuel for subsequent transmutation, will increase the radiation dose levels of the fuel. Knowledge of these dose levels could allow cost penalties associated with the handling of these materials to be estimated.

In this paper, an indication of radiation dose levels encountered in the fabrication and discharge of MA-containing fuels, irradiated in a fast reactor (FR), is presented.

MA-CONTAINING FUEL FOR A FAST REACTOR

The MA recycling concept in FR is being studied through a series of fuels containing mixed actinide oxides. They have been fabricated at the Institute for Transuranium Elements (4) and irradiated together with CEA, France in the PHENIX power station. They are currently undergoing Post-Irradiation Examinations to study their mechanical, chemical and neutronic behaviour following their discharge from PHENIX (5,6).

Four types of mixed oxide fuels (SF) were prepared in accordance with the homogeneous (SF13, SF16) and heterogeneous (SF14, SF15) concepts and irradiated in PHENIX. The specifications of the fuels are listed in Table I (4). The final burnup achieved for each fuel (Table I), was calculated on the basis of the ^{148}Nd measurement by Isotope Dilution Mass Spectrometry (IDMS).

TABLE I
Experiment SUPERFACT: Fuel and Irradiation Parameters

Fuel	Fuel type	Burn-up at %	Theo.dens. kg/m^3	Fuel.dens. % th.d.	Fuel length (m)
SF13	(0-74U 024Pu 0.02Np)02 Unat Pu238: 1.3% Pu239: 60.4% Pu240% 23.4% Pu241: 10.4% Pu242: 4.5%	6.4	11050	95	0.85
SF14	(0.6U 0.20Np 0.20Am)02 U 235: 0.3%	4.5	10970	94	0.4
SF15	(0.55U 0.45Np)02 U 235: 0.3%	4.5	11030	95	0.4
SF16	(0.74U 0.24Pu 0.02Am)02 Unat, Pu: idem SF13	6.5	11050	95	0.85
Pellet diameter: 0.00524m Pellet height: 0.007m Stainless steel cladding O.D: 0.00655m I.D: 0.00565m		Irradiation conditions: Sf13,SF16: 35000W/m SF14,SF15: 20000W/m (BOL) to 33000W/m (EOL) Irradiation time: 360 days			

THEORETICAL

Theoretical calculations were performed to obtain the neutron and gamma outputs from the four SF fuels at charge, discharge, and 1y and 5y cooling time. Calculations were also carried out on the neutron and gamma output from a standard FR fuel (25% Pu, 62 GWd/t) for comparison purposes. The MOX fuel contained plutonium of the same isotopic composition as the SF fuels. The computer code KORIGEN (7) was used for the calculations, with the fission and capture cross-sections adjusted to the PHENIX reactor. The variation of the neutron and gamma outputs for the spent fuels are shown in Figs. 1 & 2.

The two fuels containing 20% and 2% ^{241}Am respectively have the highest neutron outputs at discharge, despite having the lower burnup. These fuels achieve between one and two orders of magnitude higher neutron output than the standard FR-UPu fuel. The fuel containing 2% ^{237}Np has a neutron output of the same order of magnitude, although still higher, as the FR-UPu fuel. The 45% ^{237}Np fuel presents the lowest neutron output due to lack of plutonium at charge and consequently lower production of curium isotopes. In general,

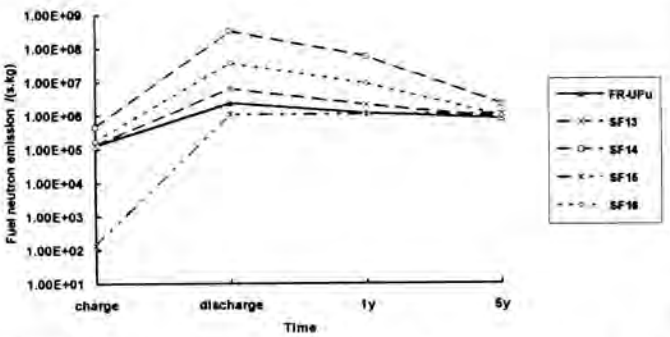

Fig. 1. Calculated neutron emission as a function of irradiation and cooling times.

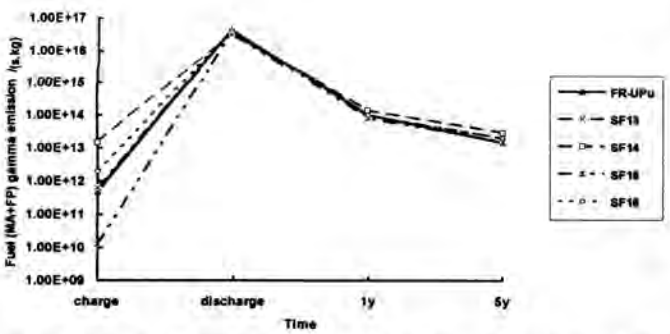

Fig. 2. Calculated gamma emission as a function of irradiation and cooling times.

^{238}Pu, ^{242}Cm and ^{244}Cm dominate the neutron emission with the curium isotopes being the main radiation sources of the americium fuels.

The gamma output at charge is higher for the fuels containing americium. At discharge, both fission products and actinides contribute to the gamma output. Small differences are observed in the gamma output. The lower fission product component due to lower burnup is compensated by the higher americium contribution in the fuels SF14 and SF16. Although there are significant reductions of the gamma emission with increasing cooling time, a weighted average energy of the gamma spectrum of about 0.5 MeV is observed throughout the cooling of the fuel.

EXPERIMENTAL

Fresh fuels containing, in particular americium, raise handling problems because of their associated gamma radiation. High neutron dose rates from curium production are also to be expected from irradiated mixed actinide oxides.

Measurements During Fabrication

The surface dose rate of the SF fuel pins containing plutonium is highly dependent on the isotopic composition. The major part of the dose rate of freshly purified plutonium from a PWR stems from neutron emission: (α, n) reactions caused by the decay of ^{238}Pu and spontaneous fission of ^{240}Pu. The dose rate from plutonium is dominated, with increasing decay time, by the gamma dose rate due to build-up of ^{237}U, and particularly, ^{241}Am from the decay of ^{241}Pu.

The gamma dose rate of the four SF fuel pins were measured during fabrication (8). The measurements were made using a portable ionisation chamber type 'Babyline' and thermoluminescence dosimeters in contact.

Measurements After Irradiation

A qualitative analysis of the FP content of the irradiated mixed actinide oxides was performed by gamma-spectroscopy. The gamma and neutron dose rates after irradiation (cooling time 57 months), were measured inside a β-γ hot cell at the Institute for Transuranium Elements. A compact array of neutron detectors and a Geiger-Müller (GM) counter embedded in a polyethylene moderator, have been used for passive neutron and gamma interrogation of the fuel (9).The gamma dose rate was measured directly, with an error of 5%, using the GM counter appropriately calibrated. The setup under which the measurements were made includes lead of 0.035m thickness between the fuel and the detector which are 0.06m apart. The neutron dose rate is obtained following the measurement of the neutron emission from the fuel through spontaneous fission and (α, n) reactions. The neutron emission is then compared to the dose rate at 1m from a ^{252}Cf neutron source of known activity.

RESULTS AND DISCUSSION

MA-containing fuels when compared to a standard UPu MOX FR fuel will generally show a higher gamma and neutron dose for the fresh and irradiated fuel. In particular, the addition of ^{241}Am increases the gamma dose of the fresh fuel and the neutron dose of the spent fuel. The ^{242}Cm and later ^{244}Cm isotopes are the strongest neutron sources for the americium containing fuel while ^{238}Pu dominates the neutron output of the $(Np_{0.45}U_{0.55})O_2$ fuel. The change in the radiation dose rate of fresh and spent MA-containing fuels in

comparison to the standard FR fuel, can be assessed by comparing in a first approach the source term nuclides which contribute to the gamma and neutron dose. The KORIGEN gamma and neutron outputs for fresh and spent fuels (5 years cooling time), normalised to the standard UPu FR fuel with the same irradiation exposure, are shown in Table II.

The KORIGEN calculations shown in Figs. 1&2, demonstrate the differences in dose rates between the standard and MA-containing fuels. In order to check the reliability of the KORIGEN predictions, the experimental and theoretical concentrations for the source term nuclides are compared (Tables III and IV). The agreement is within 25% for the actinides and 50% for the fission products, sufficient for dose rates calculations but unsuitable for neutron physics aspects. The limitations of KORIGEN with respect to its fission yield and cross-section libraries should be considered when irradiation calculations are performed. The neutronic calculations will be taken up in a separate study. It should be noted here that ^{238}Pu formed from ^{237}Np seems to be underestimated while that from ^{241}Am capture and ^{242}Cm decay overestimated.

The gamma dose rates during fabrication are given in Table V. The high radiation doses from the americium containing mixtures can be seen in the results which are consistent with those in Fig. 2 (dose rates at charge). The neutron and gamma dose rates obtained experimentally for the SF fuel pins after discharge and 57 months cooling are shown in Table VI. The total radiation dose is determined by the gamma dose rate. Even for the SF14 fuel which contains 20% ^{241}Am and for which most of the curium isotopes are expected to be produced, the gamma dose dominates over the dose due to neutrons.

TABLE II
Calculated Neutron and Gamma Outputs from the SF Fuels Normalized to a Standard FR-UPu Fuel (Spent Fuel Cooling Time 57 Months)

Fuel	Neutrons		Gamma	
	fresh fuel	spent fuel	fresh fuel	spent fuel
SF13	1	1.07	1.3	1.4
SF14	3.4	2.7	30	2
SF15	1.10E-03	1.4	0.03	1.07
SF16	1.3	1.2	4	1.4

TABLE III
Experimental and Theoretical %IMA Values

	SF13 2%Np		SF15 45%Np		SF16 2%Am	
	%IMA (exp)	%IMA (th)	%IMA (exp)	%IMA (th)	%IMA (exp)	%IMA (th)
PU238	0.68	0.54	8.4	7.6	0.57	0.52
240	6.03	5.73	0.1	0.11	5.93	5.6
Am241	0.61	0.57	-	-	1.83	1.8
Cm242	-	-	-	-	0.01	0.001
244	-	-	-	-	0.006	0.005

In conclusion, one can state that a cost penalty due to high radiation doses will be observed mainly during fuel fabrication whereas the gamma radiation dose for the spent fuel increases by less than a factor of three even in the case of the heterogeneous (Am, Np) fuel. Consequently, additional means in order to reduce the occupational hazard dose from MA-containing fuels would be required only during fabrication.

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TABLE IV
Experimental to Theoretical FP Concentrations (Normalized to Cs-137)

	SF13	SF14	SF15	SF16
Sb125	0.7	1.02	0.6	0.7
Cs134	1.8	1.6	2.1	2
Ce144	1.6	-	1.4	1.7
Eu154	2.1	2.4	1.6	2.1
155	1.4	-	0.4	0.4

TABLE V
Gamma Dose Rates (μGyls) Measured on Fuel Pins and Pellets Without Shielding During Fabrication

	SF13	SF14	SF15	SF16
1 pellet (1.8E-3 kg)				
contact	0.02	0.83	0.03	0.08
Fuel Pin:				
contact	0.12	5	0.4	0.5
0.5m distance	5.00E-03	0.25	0.01	0.03

TABLE VI
Radiation Dose Rates of Spent Fuel (Cooling Time 57 Months)

Fuel	Neutron Dose Rate nSv/(s.kg)	Gamma Dose Rate mGy/(s.kg)
SF13	2.6	n.m.
SF14	5.5	0.5
SF15	3.1	0.4
SF16	2.6	1.4

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