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THE US/UK ACTINIDES EXPERIMENT AT THE DOUNREAY PFR

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DISCLAIMER

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ABSTRACT

The United States and the United Kingdom have been engaged in a joint research program in which samples of higher actinides were irradiated in the 600-MW Dounreay Prototype Fast Reactor in Scotland. Analytical results using mass spectrometry and radiometry for actinides and fission products are now available for the samples in Fuel Pins 1 and 2, which were irradiated for 63 full-power days, and for the samples in Fuel Pin 4, which were irradiated for 492 full-power days. Results from these three fuel pins are providing estimates of integral cross sections and fission yields.

I. INTRODUCTION

A joint venture between the United States and the United Kingdom to investigate the physics of higher actinides exposed in a fast reactor has been in operation since 1979. Objectives of the cooperative program are (a) to secure improved knowledge of the basic nuclear cross sections and reaction rates, (b) to provide data for assessment of the fuel worth and alternative waste management options, and (c) to obtain a preliminary evaluation of higher actinide oxides in a fuel-type irradiation.

Four irradiation-capsule pins were prepared[1] which had the outward appearance of standard fuel pins in use at the Dounreay Prototype Fast Reactor (PFR). Three of these pins contained a series of encapsulated actinide isotopes in milligram quantities.[2–4] There were altogether 21 individual actinides ranging from ^{230}Th to ^{248}Cm as follows:

Thorium: ^{230}Th, ^{232}Th

Protactinium: ^{231}Pa

Uranium: ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U

Neptunium: ^{237}Np

Plutonium: ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{244}Pu

Americium: ^{241}Am, ^{243}Am

Curium: ^{243}Cm, ^{244}Cm, ^{246}Cm, ^{248}Cm

There was an encapsulated sample for each one of these actinide species (and two samples for a few species). Most of the samples also contained minor amounts of other related actinide species. Each sample, however, is identified with one of the above species which was the principal actinide in that sample.

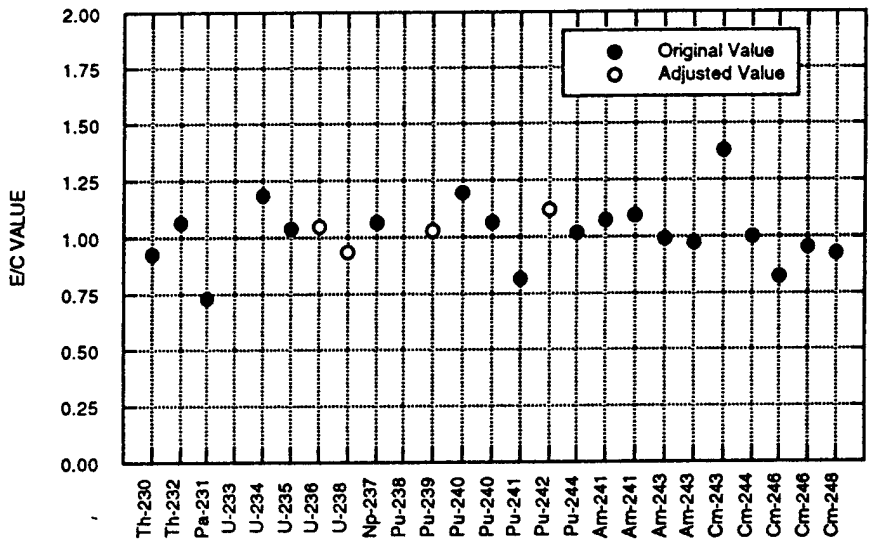

Figure 1. E/C ratio for the primary actinide concentration. This figure and supporting data are discussed in greater detail in Ref. 15. The adjustments to the E/C values are also discussed there.

II. ANALYSIS AND RESULTS

Fuel pins 1 and 2 were irradiated for 63 full-power days. The laboratory analysis of the samples contained in FP-1 and FP-2 was performed at Oak Ridge National Laboratory (ORNL) and has been discussed in previous reports.[5–8] A third pin, FP-3, was also irradiated for 63 full-power days. It was sent to AERE Harwell and shall not concern us further. The fourth pin (FP-4) was the most extensively irradiated (equivalent to 492 full-power days) of these four pins. The PFR power history during the irradiation process is given in Ref. 9. For the total exposure experienced by FP-4, the neutron fluence was approximately 2×10^{23} n/cm^2. Analyses of the FP-4 samples (also performed at ORNL) have now been completed.[10] Mass and alpha-spectrometry were used to measure the actinide concentrations, and gamma-ray spectrometry was used for the fission products. The fission products analyzed were ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{152}Eu, ^{154}Eu, and ^{155}Eu.

Time-dependent neutron flux spectra were supplied by the UK for the sample locations in the PFR core, and these, together with ENDF/B-V cross-section and ENDF/B-VI fission-yield data, were used to develop one-group cross sections and fission yields. These cross sections and yields were used with the ORNL code ORIGEN-S [Ref. 11] to simulate the irradiation process and predict actinide transmutation rates and fission-product production. Preliminary comparisons have been made between the measured and the calculated reaction products.[12–16] This analysis showed some results where the Experiment to Calculated (E/C) ratio for actinide amounts following irradiation was significantly different from unity. Some of this could be attributed to difficulties encountered during laboratory analysis. It was possible via inter-comparisons among the samples using other actinide species contained in the samples together with quite accurate values of isotopic ratios to identify samples where adjustments to the measured values could be estimated. The same adjustments were then applied to the fission-product data.

The general trend in the actinide results is well illustrated by Fig. 1 which shows the E/C values for the principal actinides. These values are reasonably close to unity. From the fission-product data, the results for ^{137}Cs are summarized in Fig. 2 which shows the E/C values for each of the samples identified by its primary component. The values for most of the plutonium, americium, and curium isotopes are reasonably close to, if somewhat lower than, unity. For the thorium, protactinium, uranium, and neptunium isotopes, however, the fission-product results are rather variable. We are currently attempting to better understand these actinide and fission-product results.

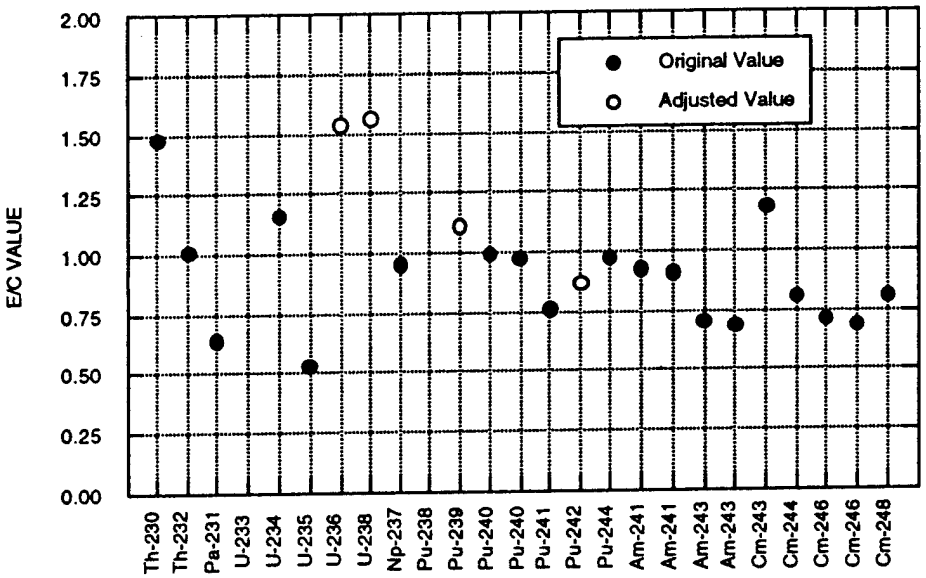

Figure 2. E/C ratio for ^{137}Cs in each of the actinide samples versus the principal fissioning actinide. These are the results following adjustments indicated from an analysis of the actinide data. See Ref. 15 for details.

III. ACKNOWLEDGMENTS

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(19) UC-900, DOE