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US/UK ACTINIDES EXPERIMENT AT THE DOUNREAY PFR
I. FISSION PRODUCTS

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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.

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.

ANALYSIS

Fuel pins 1, 2, and 3 were irradiated for 63 full-power days. The laboratory analysis of the FP-1 and FP-2 samples was performed at Oak Ridge National Laboratory (ORNL) and has been discussed in previous reports *(5-8)*. The third pin (FP-3) 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. Figure 1 shows the PFR power history during the irradiation process *(9)*. For the total exposure indicated in Fig. 1 (i.e., that experienced by FP-4) the neutron fluence was approximately 2.0×10^{23}. 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.

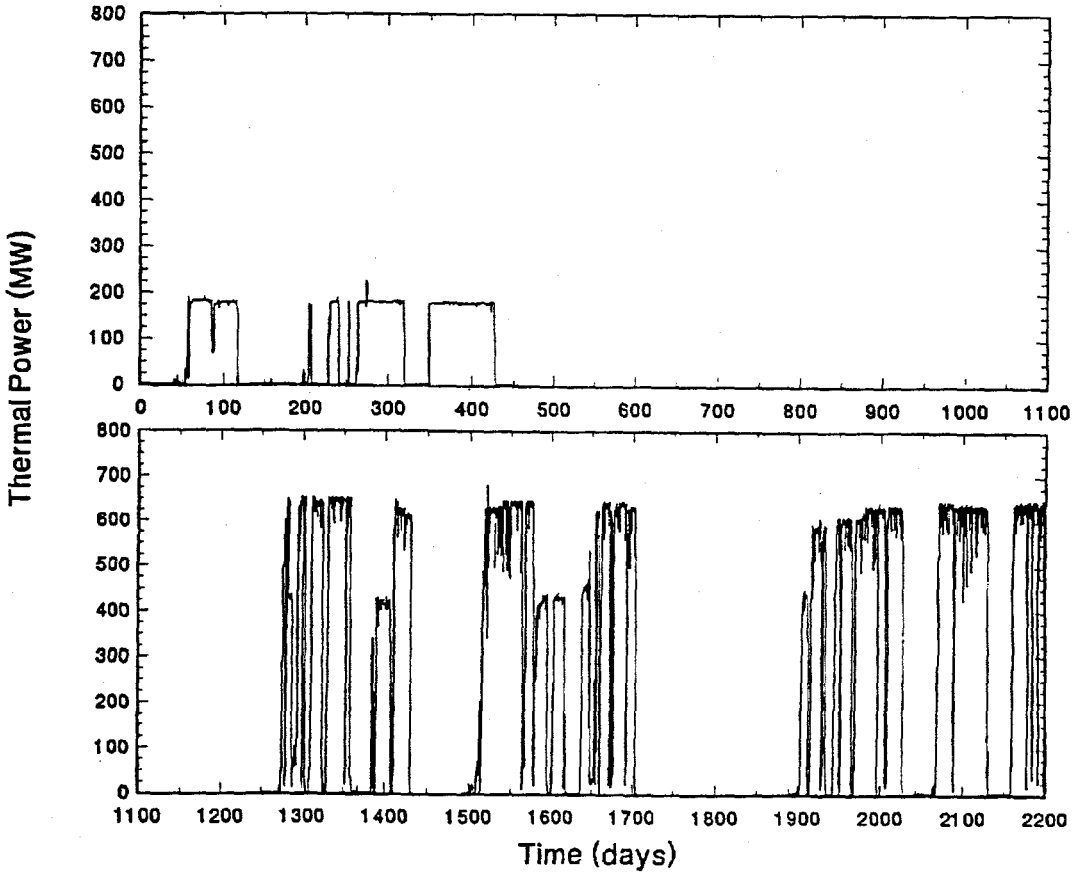

Figure 1: Power history during the exposure of the actinide samples. The experiment occurred over a 2200-day time period. FP-1 and FP-2 were exposed for the first 470 days (see top panel). FP-4 was exposed for the full 2200 days.

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 *(11)* to simulate the irradiation process and predict actinide transmutation rates and fission-product amounts. Preliminary comparisons have been made between the measured and the calculated reaction products *(12,13)*.

OVERVIEW OF RESULTS

We present here some detailed results with particular emphasis on the fission products. We show comparisons between experimentally measured and calculated values. In general, the results show reasonable agreement between measured and calculated values. The radiochemical analysis required in this set of experiments was very demanding. All actinide material in a sample, including the vanadium used for encapsulation, was first dissolved in nitric acid. Dissolution proved to be a slow process for some of the actinides and boiling of the solution was generally required. For some samples, material was lost through spillage of the boiling solution. By comparisons among samples and by noting the amount of other actinides present in a sample and, in turn, by comparing with other samples containing those same actinides, and by comparison with dosimeter samples, it was possible in some cases to correct for lost sample material.

As regards the primary actinide in each sample, one can examine the amount following irradiation or, alternatively, one can examine the amount of primary actinide that is depleted. Ideally, the latter is a more useful quantity. However, when cross sections are low and/or when sample sizes are small, the measured values for this quantity will be subject to large uncertainty. For a majority of the actinides, experimental values for the amount of actinide remaining following irradiation, and for the amount of one- and two-neutron capture products generated, compare reasonably with predictions. There are some differences in calculated values depending on whether one uses U.S. or U.K. cross-section data sets *(14)*. Work is continuing on the actinide results.

FISSION-PRODUCT RESULTS

The general trend in the fission-product results is well illustrated by Fig. 2 which shows the experimental-to-calculated ratio (E/C) for the ^{137}Cs content of the exposed samples. Results for all three pins analyzed at ORNL are included. These data are for all samples that yielded reliable ^{137}Cs results. Samples that were damaged, lost in their entirety, or where loss occurred that could not be corrected for, are not included. The results for FP-1 and FP-2 are from Tables 13 and 14 of Ref. *(7)*. Uncertainties are not shown here but are discussed in Refs. *(6)* and *(7)*. The FP-4 ^{137}Cs results were first reported by Murphy *et al. (12)* and as reported here they contain some minor updates. The calculated values in the FP-4 case used fission yields as compiled by Brady *et al. (15)* Samples are identified by the primary or principal fissioning actinide. In Fig. 2 there are 19 principal actinides (there are two samples each of ^{240}Pu, ^{241}Am and ^{243}Am). Of the 21 principal actinides exposed and that were listed above, neither ^{238}Pu nor ^{243}Cm yielded reliable results.

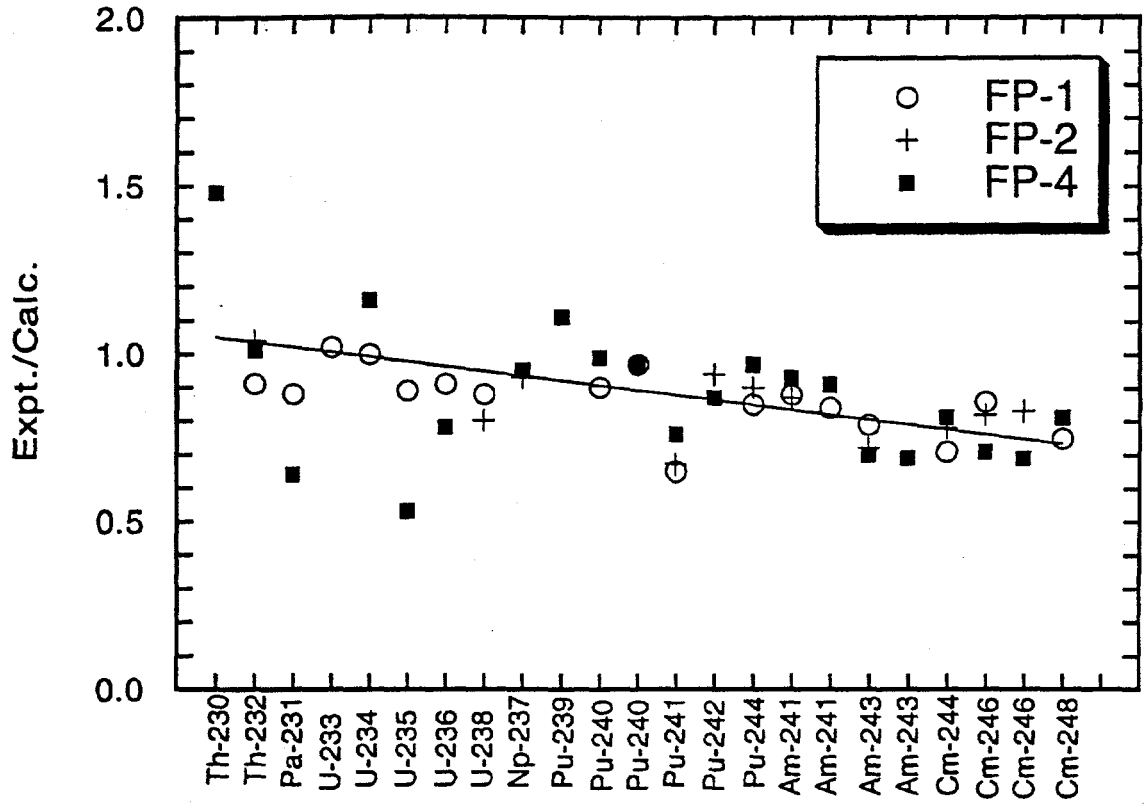

Figure 2: Experimental-to-calculated (E/C) ratios for ^{137}Cs concentrations in FP-1, FP-2, and FP-4. All samples that yielded reliable results are included. A line is drawn to guide the eye. This line shows a steady downward trend in E/C values.

In general, there is agreement between the results obtained from the three fuel pins. This is particularly so for the plutonium, americium and curium isotopes. The FP-4 results for ^{230}Th, ^{231}Pa and ^{235}U are poor. The ^{230}Th sample was small and the fission cross section is low, so the uncertainty is large. When the ^{231}Pa results from FP-4 are examined together with the actinide results from the same sample it seems that there may have been some loss of material. The ^{235}U result is disturbing. One would expect better agreement for this sample. The ^{134}Cs result for the ^{235}U sample is also low although some of the other fission-product measurements in this sample give E/C values close to unity. Thus, there may be a problem involving the cesium chemistry for this particular sample.

There seems to be a steady downward trend in E/C for the americium and curium samples (see Fig. 2). One must keep in mind that these E/C values are not simple measures of the agreement between experimental and theoretical fission yields. They are comparisons between measured and predicted ^{137}Cs concentrations in the samples. The ^{137}Cs concentration results from the fissioning of *all* species present in the sample and includes contributions from the various minor actinides. One must further take account of the fissioning of species that accumulate in the samples via neutron capture and related reaction pathways. In the case of FP-4, which was exposed to a much greater neutron fluence, the influence of neutron-capture products was much more in evidence. Nevertheless, it does seem that the evaluated fission yields for some of the americium and curium isotopes are in need of revision.

The results shown in Fig. 2 for ^{137}Cs are typical of what one sees for most of the other fission products. There are, however, two exceptions: ^{106}Ru and ^{110m}Ag. As an illustration, we refer to Fig. 3 which is a plot of the E/C for all nine fission products in the case of five of the plutonium samples in FP-4. The trend seen in Fig. 3 is typical of what one sees in the other samples of FP-4 when all nine fission products are examined. Specifically, E/C values are low for ^{106}Ru, they are high for ^{110m}Ag, and they are reasonable for the other seven fission products. Both the low value for ^{106}Ru and the high value for ^{110m}Ag are evident in all the actinide samples of FP-4.

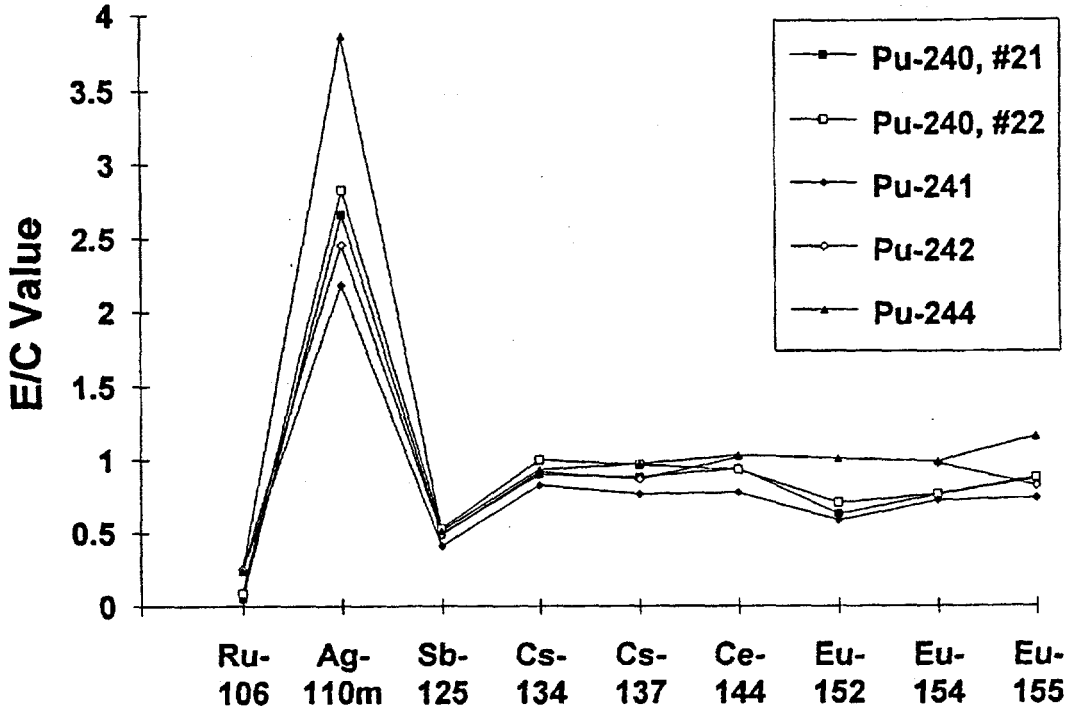

Figure 3: Experimental-to-calculated (E/C) ratios for the nine fission products in five plutonium samples. This general pattern is also seen for the other samples in FP-4.

We note that ^{110m}Ag is a shielded nuclide (so also are ^{134}Cs, ^{152}Eu, and ^{154}Eu). Most of the ^{110m}Ag concentration will result from neutron-capture reactions on ^{109}Ag rather than being the direct result of fission (the cumulative fission yield is much greater for ^{109}Ag, which now acts as target material, than is the independent fission yield of ^{110m}Ag). What is likely the case is that the ^{109}Ag neutron-capture cross section used in the calculations is too low. The consistently low E/C values obtained for ^{106}Ru seem to be explained by the fact that ruthenium evaporated during dissolution *(16)*.

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(16) A similar conclusion was reached independently by JAERI scientists who made fission-product measurements on samples that were provided to them for cross-checking purposes. K. Gunji, private communication.