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NEUTRONICS BENCHMARK OF A MOX ASSEMBLY WITH NEAR-WEAPONS-GRADE PLUTONIUM

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I. INTRODUCTION

One of the proposed ways to dispose of surplus weapons-grade plutonium (Pu) is to irradiate the high-fissile material in light-water reactors in order to reduce the Pu enrichment to the level of spent fuels from commercial reactors. Considerable experience has been accumulated about the behavior of mixed-oxide (MOX) uranium and plutonium fuels for plutonium recycling in commercial reactors, but the experience is related to Pu enrichments typical of spent fuels quite below the values of weapons-grade plutonium. Important decisions related to the kind of reactors to be used for the disposition of the plutonium are going to be based on calculations, so the validation of computational algorithms related to all aspects of the fuel cycle (power distributions, isotopics as function of the burnup, etc.), for weapons-grade isotopics is very important.

Analysis of public domain data (see Ref. 1) reveals that the cycle-2 irradiation in the Quad Cities boiling-water reactor (BWR) is the most recent U.S. destructive examination. This effort involved the irradiation of five MOX assemblies using 80 and 90% fissile plutonium. These benchmark data were gathered by General Electric under the sponsorship of the Electric Power Research Institute. It is emphasized, however, that "global" parameters are not the focus of this benchmark, since the five bundles containing MOX fuels did not significantly affect the overall core performance. However, since the primary objective of this work is to compare against measured post-irradiation assembly data, the term benchmark is applied here.

One important reason for performing the benchmark on Quad Cities irradiation is that the fissile blends (up to 90%) are higher than reactor-grade and, quite close to, weapons-grade isotopics.

II. DESCRIPTION OF THE MEASURED MOX ASSEMBLY

Four of the five assemblies were located at the center of the reactor; the fifth assembly was at the periphery between the core and the reflector . All were irradiated during cycle 2 (July 21, 1974, to January 2, 1976). In particular, 9 rods of the assembly named GEB-161 were destructively analyzed after the end of cycle 2 at different axial positions. Because of a lack of details about the void fraction profile we compare results at the first measured axial location [elevation 53.34 cm (21 in.) from inlet] with a zero void. Figure 1 describes the 7×7 bundle and the location of the 9 rods that are the subject of this intercomparison. Details about compositions, geometry and irradiation history were taken from Ref. 2. The average burnup for the central MOX assemblies is quoted as 8300 MWd/tonne (717.12 MJ/g), with a particular value of 11,206 MWd/tonne (968.20 MJ/g) for assembly GEB-161 at the elevation of 53.34 cm (21 in.). Of the 9 measured rods, 4 contained UO_2 fuel, 4 MOX fuel and 1 contained UO_2 with gadolinium as the burnable poison. Tables 1 and 2 contain isotopic details, and Table 3 contains geometric details.

Table 1. Composition of Assembly GEB-161[a]

Rod Type	Number	U-235 wt%	Fissile Pu wt%	Gd2O3 wt%	Density g/cc
1	8	2.56	---	---	10.32
2	9	1.94	---	---	10.32
3	6	1.69	---	---	10.32
4	1	1.33	---	---	10.32
5	10	3.30	---	---	10.32
6	4	2.56	---	3.0	10.19
7	1	2.56	---	2.5	10.19
P1 (solid)[b]	2	0.72	2.14	---	9.89
P2 (solid)[c]	3	0.72	3.52	---	9.89
P3 (annuli)[b]	2	0.72	2.34	---	8.94
P4 (annuli)[c]	3	0.72	3.62	---	8.94

[a]Zircaloy-2 for clad, Zircaloy-4 for box (density 6.55 g/cc).Water density, 0.7375 g/cc.
[b]80% (relative to Pu) fissile. Fissile Pu is the weight of Pu-239 and Pu-241 divided by the total heavy-metal mass.
[c]90% (relative to Pu) fissile. Fissile Pu is the weight of Pu-239 and Pu-241 divided by the total heavy-metal mass.

Table 2. Plutonium Isotopics in MOX Fuel (wt %)

Nominal	Real	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
80	80.13	0.25	75.66	18.49	4.47	1.13
90	89.54	0.12	87.16	10.06	2.38	0.28

Table 3. Geometry of Assembly GE-161

Magnitude	Dimension (cm)
Corner Radius	1.079
Box Width	13.813
Box Thickness	0.203
Narrow/Narrow Ga	0.950
Wide/Wide Gap Thickness	1.905
Lattice Pitch	1.874
Clad Outside Radius	0.716
Clad Inside Radius (UO_2)	0.622
Clad Inside Radius (MOX)	0.635
Pellet Radius (UO_2)	0.607
Pellet Radius (MOX)	0.620
Pellet Inside Diameter (MOX annuli)	0.381

III. MODELING OF THE ASSEMBLY GEB-161

The information in Tables 1 to 3 is the input data for the two-dimensional (2-D) code HELIOS, Ref. 3, resulting in a description of the system identical to what is shown in Fig. 1. HELIOS, a Scandpower, Inc. proprietary code whose license was purchased by ORNL, uses the collision probability methodology to calculate the transport of neutrons and photons in 2-D geometries. Radial details were included for the pins with burnable poisons in order to better simulate the gradual burning of gadolinium. The input value for the average burnup of the 49 pins (11,772 Mwd/tonne, 1017.10 MJ/g) was iterated until a value of 11,890 Mwd/tonne (1027.30 MJ/g, a value from Ref. 2) was obtained for the average burnup of the 9 destructively analyzed pins. Fourteen equally spaced burnup steps were computed along the 18-month irradiation period. A white reflective boundary condition was imposed at the middle of the water channels.

The output of the HELIOS calculation was limited intentionally since only the following available experimental data are useful for comparison purposes: (1) gadolinium isotopics for the burnable poison pin; (2) isotopics of uranium, plutonium, and neodymiun; (3) number densities of Np-237, Am-241 and Cm-244; (4) relative isotopics of Cm-242 and (Cm-243 + Cm-244) and Am-241, Am-242 and Am-243; (5) burnup distribution between the pins; and (6) relative power distribution between the pins averaged during the last few weeks of the irradiation via the activity of the fission product La-140 (mainly the daughter of Ba-140, which has a half-life of 12.75 days).

IV. COMPARISON BETWEEN CALCULATED AND EXPERIMENTAL VALUES (C/E)

A considerable amount of information is related to this benchmark. A summary is presented in Fig. 2, where the average results for the four UO_2 pins and the four MOX pins are shown. The experimental errors indicated for the density of each isotope are the combinations of the errors summarized in Ref. 2. After considerations of how the measurements were presented we determined four categories: (1) atomic densities relative to original U-238 atomic densities for U, Pu and Nd isotopes; (2) atomic densities per milligram of original U-238 for Np-237, Am-241 and Cm-242; (3) isotopic fractions of Cm-242 and (Cm-243 + Cm-244); and (4) isotopic fractions of Am-241, 242 and 243.

Comparing C/E values with the experimental errors we conclude:

1. Uranium isotopes: accurately calculated values for U-235 and U-238 concentrations in both types of pins; calculated U-236 concentrations are within experimental error for MOX pins but are very low for UO_2 pins.

2. The calculated Np-237 concentrations are within the large experimental error, but systematically smaller.

3. Plutonium isotopes: there are systematically better values of C/E for the MOX pins in comparison to the UO_2 pins. Concentrations of the important isotope 239 are smaller by 3% and 6.5% with respect to the high-accuracy measurement in MOX and UO_2 pins, respectively. Results are within experimental error for the concentrations of Pu-240 and Pu-241 in the MOX pins.

4. Americium isotopes: the very large experimental errors are related to the absolute measurement of Am-241 (30% and 100% errors for MOX and UO_2 pins, respectively) and the measurement of the isotopic fractions of Am-243 (2.8% and 68% for MOX and UO_2 pins, respectively). The large experimental errors make inconclusive the comparison, with the exception of the detection of very small values for the concentration of Am-241 in the MOX pins.

5. Curium isotopes: in general, calculated values are systematically smaller than experimental values, although compatible with experimental errors. The exception is that the MOX pins show very small values for the Cm-242 concentrations.

6. Neodymium isotopes: measurement and calculations look, in general, compatible with somewhat better agreement for the MOX pins.

7. Because of the normalization of the calculations the average burnup values for the nine pins have a C/E = 1. More meaningful, the relative distribution, not shown in Fig. 1, is consistent with experimental values within a few percent.

Because there are direct measurements of the relative (with respect to the element) isotopic fractions of Cm-242 and Am-243 we compared them with calculated values. In general the results for the UO_2 pins are better than the MOX pins for both isotopic fractions.

V. CONCLUSIONS

The work presented here provides valuable assembly level comparisons for a MOX and UO_2 assembly under reactor operating conditions. The experience gained leads to the following conclusions:

- For MOX rods, HELIOS models the chains for the isotopes of uranium and plutonium reasonably well compared with measured data at approximately 12,000 Mwd/tonne (1036.80 MJ/g). At higher points in the transmutation chains, the calculational and experimental difficulties make the comparison more uncertain. This is particularly important for the case of Cm isotopes since these neutron sources may be an important factor in shielding analysis.

- The nondestructive pin power measurements are in good agreement with the calculated values. This includes the gadolinium pin.

- Burnup distributions between the measured pins show consistency between experimental and calculated values.

- The analysis presented here provides a reasonable starting point and a necessary first step toward the validation of neutronics methods for high-fissile MOX fuels. A review of the Quad Cities data suggests that more benchmarks are possible after including void fraction values in the calculations.

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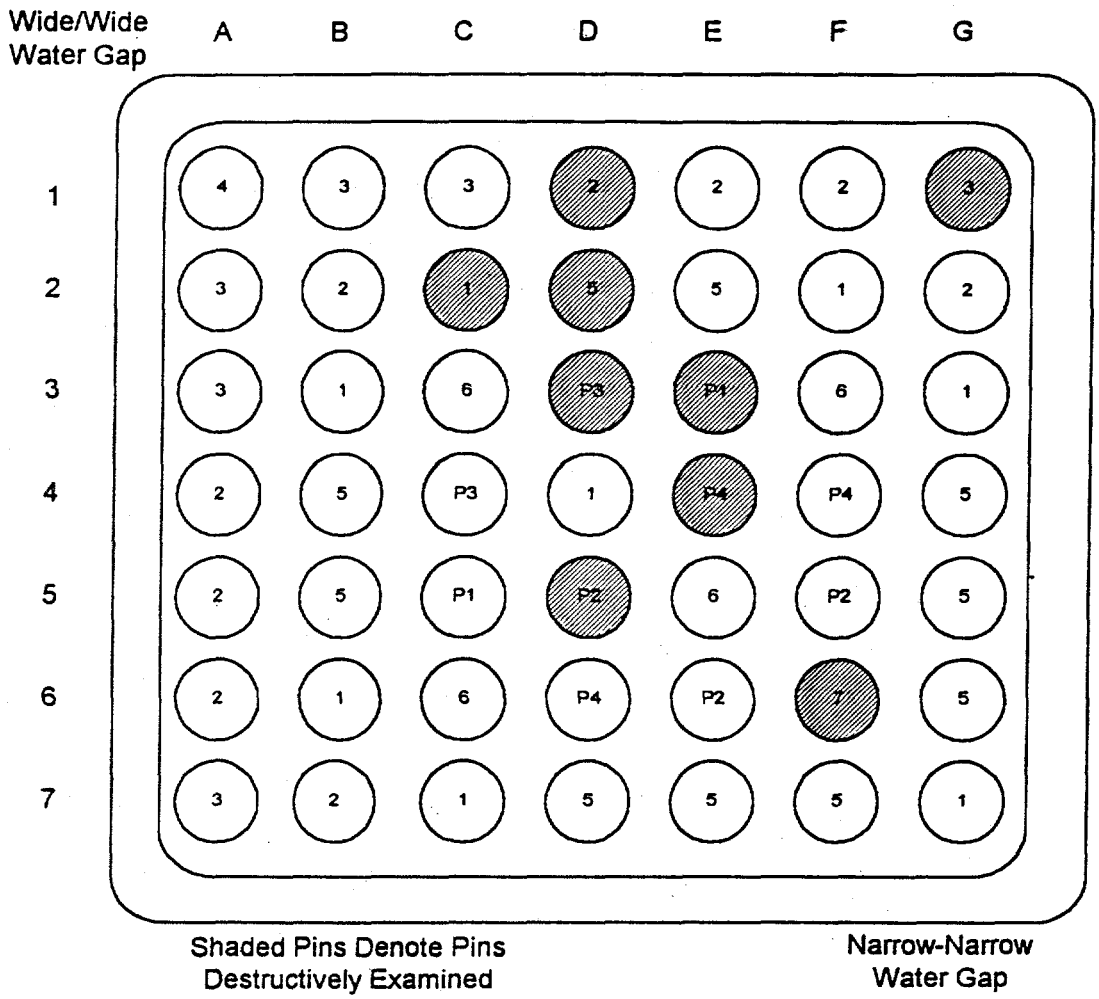

Fig. 1. Quad cities MOX central assembly GEB-161. P1–P4 pins are the highly enriched plutonium MOX fuels; the number refers to types of fuels described in Table 1.

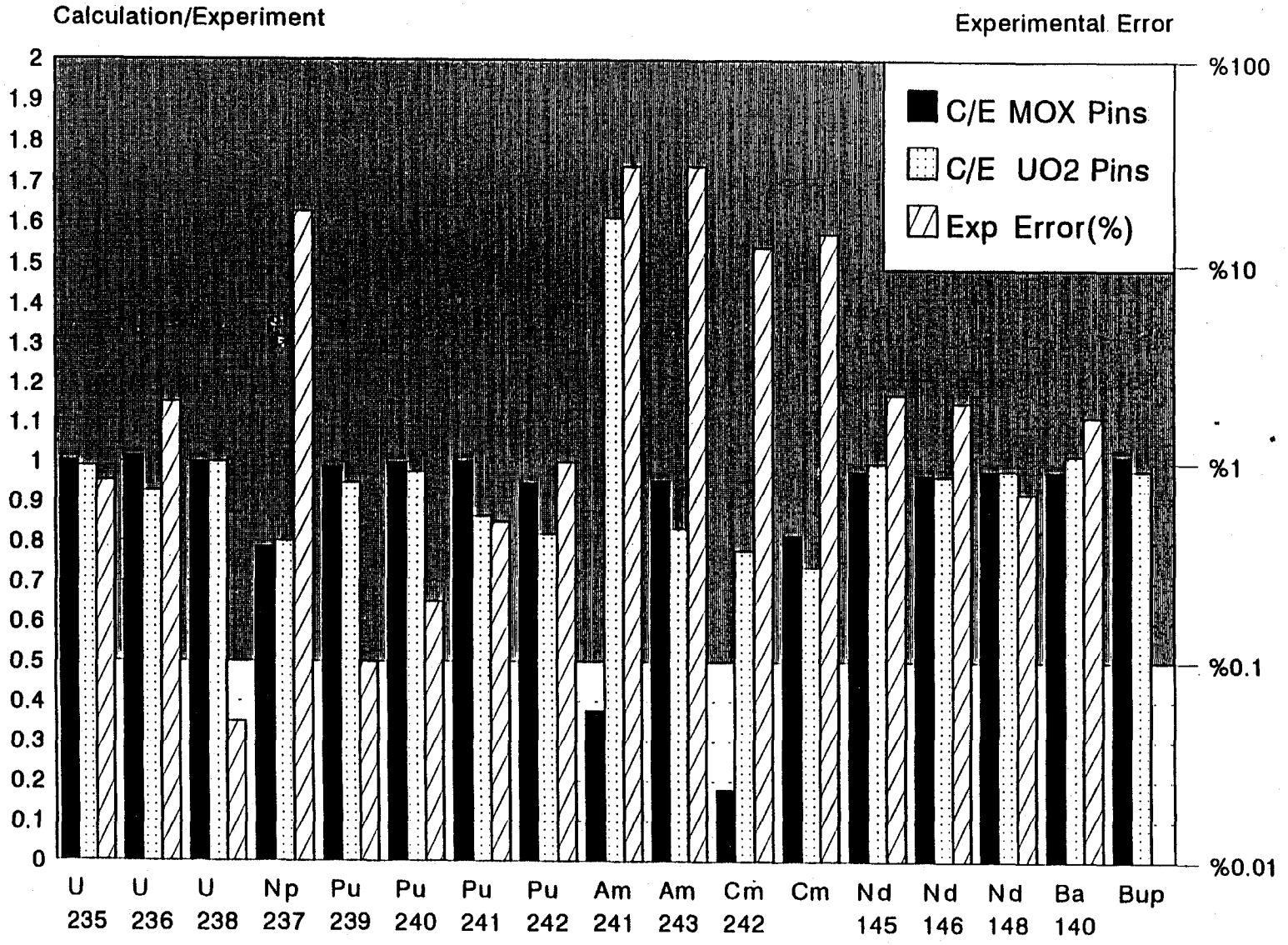

Fig. 2. Calculated/experimental ratios for the average number densities, power (Ba-140) and burnup of four UO_2 and four MOX pins of assembly GEB-161. Cm = Cm-243 + Cm-244.