Evaluation of Water-Moderated Lattices of UO_2-2.0 wt. % PuO_2 Rods

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Abstract

In the mid 1960's, three sets of experiments with lattices of mixed oxide (2 wt. % PuO_2) fuel rods were performed at the Critical Approach Facility at Hanford Operations. These experiments have been evaluated as part of the International Criticality Safety Benchmark Evaluation Project for use by criticality safety specialists. This paper presents a description of the experimental configuration and discusses the benchmark model developed for the experiments. A sensitivity analysis determining the effects of data uncertainties on the k_{eff} value calculated from the benchmark model is described. The k_{eff} values calculated using the benchmark model with both MCNP, with continuous-energy ENDF/B-V cross section data, and KENO-Va, with 27 group ENDF/B-IV cross sections, are given. Trends observed in the calculated k_{eff} values and possible conclusions, which may be drawn from these trends, are discussed.

I. Introduction

In the mid 1960's, three sets of experiments involving mixed oxide fuel rods were performed at the Critical Approach Facility at Hanford Operations in Richland, Washington. Uotinen et al[1] reported the results of these experiments in 1972. As part of the International Criticality Safety Benchmark Evaluation Project (ICSBEP), these experiments have been analyzed to provide benchmark descriptions for use by nuclear criticality safety specialists to validate computational codes and cross sections. This paper presents an evaluation of these experiments.

The development of a benchmark model includes developing a simplified model of the experiments from the available data and performing a sensitivity analysis on physical parameters associated with the benchmark model. A sensitivity analysis is performed to

determine the effect of uncertainty associated with the available data on the calculated multiplication factor (k_{eff}). Also, physical parameters, which may affect k_{eff} but cannot be included in the benchmark model, are evaluated.

Section II of this paper describes the configuration of the experiments, including geometry data, material data, and the experimental procedure. Section III describes the sensitivity analysis performed on the benchmark model that was developed. Section IV gives the results of k_{eff} calculations using the benchmark model for KENO-Va[2] and MCNP[3] and a brief discussion of these results.

II. Experimental Configuration

A total of seventeen experiments were performed with UO_2-2.0 wt. % PuO_2 fuel rods in triangular pitch lattices moderated by light water. Experiments were performed with six lattices containing ~8% ^{240}Pu fuel rods, five lattices containing ~16% ^{240}Pu fuel rods, and six lattices containing ~24% ^{240}Pu fuel rods. Experiments with a lattice spacing of 2.0320 cm, 2.3622 cm, 2.6670 cm, 2.9032 cm, 3.3528 cm, and 3.5204 cm were performed with the 8% and 16% ^{240}Pu rods. Lattices with the 16% ^{240}Pu rods were constructed for all of these lattice pitches except for 2.0320 cm.

An approach-to-critical technique was used to find the number of rods required to make each lattice exactly critical. Neutron multiplication data was collected at a number of loading sizes between about 50% and 96% of the critical number of rods. A straight line was fit by the method of least squares to a plot of (N/f) versus N, where N is the number of rods and f is the count rate. Extrapolation of this line to (N/f) equal to zero produced the number of rods for a critical lattice. Table I gives this experimentally determined critical number of rods for each lattice.

Table I – Critical Number of Rods

Pitch (cm)	8% ^{240}Pu Critical Size (No. Rods)	16% ^{240}Pu Critical Size (No. Rods)	24% ^{240}Pu Critical Size (No. Rods)
2.0320	319.7 ± 0.1		519.5 ± 0.1
2.3622	192.4 ± 0.1	245.6 ± 0.1	286.1 ± 0.1
2.6670	152.1 ± 0.2	194.3 ± 0.1	233.2 ± 0.1
2.9032	147.5 ± 0.1	187.5 ± 0.1	232.1 ± 0.1
3.3528	163.1 ± 0.1	221.1 ± 0.1	296.2 ± 0.1
3.5204	179.5 ± 0.2	254.6 ± 0.1	365.3 ± 0.1

Because the approach-to-critical method was used, no exact critical configuration of the fuel rods can be determined. The experimenters added rods so that the lattice was approximately a right circular cylinder at all loadings. The lattice was reflected by water but both the radial thickness and the depth of the water below the lattice were not given. However, because the lattice was reported as effectively fully reflected in all directions,

neither the exact dimensions of the reflector nor a description of the components of the room in which the experiments were performed was required.

The fuel rods were 91.44 cm in length with a diameter of 1.2827 cm. The bottom 0.5 cm of the fuel portion of each rod was a region of UO_2 powder containing natural uranium. Each fuel rod was clad with 0.0762 cm thick Zircaloy-2 and was capped by approximately cylindrical Zircaloy-2 top and bottom end plugs 0.8255 cm and 0.6985 cm long respectively. Because the fuel was vibrationally compacted, there was no gap between the fuel and the clad.

The fuel consisted of 2.0 wt. % PuO_2 mixed with UO_2 in which the uranium was natural uranium. The isotopic composition of the 8% ^{240}Pu fuel, given in Table II, was taken from Smith and Konzek[4], which describes a different set of experiments that used the same fuel rods. The measurements for these experiments were performed more recently and thus the reported material data was used instead of the fuel composition reported by Uotinen et al, which varied slightly. No other data was available for the composition of the 16% and 24% ^{240}Pu rods, so the fuel composition reported by Uotinen et al, given in Table III, was used for the experiments involving these rods. The uranium isotopic composition was not specified, but the uranium was reported to have been natural uranium, so standard isotopic ratios from reference 5 were used. Corrections were made to the material data to account for the decay of ^{241}Pu, which decays to ^{241}Am with a half-life of 14.4 years. The density of the mixed oxide region was 9.54 g/cm^3 and the density of the uranium oxide layer was 9.286 g/cm^3.

Table II – Fuel Material Composition, 8% ^{240}Pu

Isotope	Weight Percent
^{238}Pu	0.009 ± 0.002
^{239}Pu	91.835 ± 0.018
^{240}Pu	7.760 ± 0.018
^{241}Pu	0.367 ± 0.004
^{242}Pu	0.028 ± 0.002
^{241}Am	64.6 ± 0.3 ppm
^{234}U	0.0059 ± 0.0006
^{235}U	0.708 ± 0.004
^{236}U	< 10 ppm[a]
^{238}U	99.284 ± 0.004
Uranium	86.02 ± 0.06
Plutonium	1.788 ± 0.001

[a] ppm by weight of ^{236}U in UO_2-PuO_2 mixture

Table III – Fuel Material Composition, 16% ^{240}Pu and 24% ^{240}Pu

Isotope	16% ^{240}Pu (at. %)	24% ^{240}Pu (at. %)
^{239}Pu	81.11	71.76
^{240}Pu	16.54	23.50
^{241}Pu	2.15	4.08
^{242}Pu	0.20	0.66
^{234}U	0.0055	0.0055
^{235}U	0.720	0.720
^{238}U	99.2745	99.2745

The fuel rods were embedded 0.254 cm into a 1.905 cm thick lucite ($C_5H_8O_2$) support plate. Two 1.905 cm thick lucite templates 22.5425 cm and 68.8975 cm above the top of the support plate held the rods in place. A 1.27 cm thick aluminum support plate was positioned 14.9225 cm below the lucite support plate with water in between. The radial dimensions of these support structures are unknown. Three PuBe sources, each containing 80 g of Pu, were positioned 6.35 cm below the top of the lucite support plate. Also, three BF_3 neutron detectors were placed at the centerline of the fuel core at a radial distance of 40.64 cm.

III. Analysis

A benchmark model was developed for each lattice including the mixed oxide fuel, the UO_2 region, the Zircaloy-2 cladding and end plugs, the lucite support structures, the aluminum support plate, and the water reflector. A sensitivity study was performed for each lattice to determine the effects of measurement uncertainties on k_{eff}. Because no uncertainties were given for most of the data, measurements were assumed to be accurate to the smallest reported significant digit, with an uncertainty of half the smallest digit. Sensitivity studies were performed with KENO-Va with 27-group ENDF/B-IV cross sections[2]. The uncertainty in k_{eff} resulting from the uncertainty in a parameter was found by comparing the k_{eff} of the perturbed and benchmark cases (i.e. $\Delta k = k_{pert} - k_{benchmark}$).

Sensitivity analyses were performed with the following parameters: fuel length, fuel diameter, radial extent of lucite support structures, Pu wt. %, Pu isotopic composition, U isotopic composition, fuel density, clad thickness, lattice pitch, clad composition, temperature, lucite thickness, lucite density, lucite plate separation, and reported number of rods. A sensitivity study was also conducted to determine the effect of the uncertainty in the shape of the critical lattices resulting from the use of the approach-to-critical method. The 8% ^{240}Pu case also had an additional uncertainty arising from the discrepancy in the material data given in references 1 and 4. The total uncertainty in k_{eff} from measurement uncertainties was the square root of the sum of the squares of the uncertainty for each parameter. Individual parameter uncertainties and the total uncertainty in k_{eff} for each experiment were reported.

An additional uncertainty arises in k_{eff} from the use of an integral number of fuel rods for the benchmark model. The critical number of rods was reported to within a tenth of a rod, and because the number of rods was rounded to the nearest integer for the benchmark model, a bias accounting for the difference between the reported number of rods and the nearest integral number, was applied to the experimental k_{eff}. Also, an additional bias to the experimental k_{eff} is required to account for the particulate nature of the PuO_2 in the fuel. The nonhomogeneity of the mixed oxide fuel causes a reactivity defect, which was estimated by Liikala et al[6]. This estimate was used for the bias accounting for the fuel inhomogeneity. The use of this bias also presents an uncertainty in the benchmark k_{eff}. This uncertainty was conservatively assumed to be 100% of the bias.

IV. Results

MCNP with standard ENDF/B-V cross sections[7] and KENO-Va with 27-group ENDF/B-IV cross sections were used to calculate k_{eff} for the benchmark models of the seventeen lattices. The experimental k_{eff} with the biases and the k_{eff} from the benchmark calculations are given in Tables IV-VI.

Table IV – Results of Sample Calculations, 8% ^{240}Pu

Pitch (cm)	Experimental k_{eff} with biases	KENO (27-Group ENDF/B-IV) k_{eff} ± Std. Deviation	MCNP (Continuous Energy ENDF/B-V) k_{eff} ± Std. Deviation
2.0320	1.0014 ± 0.0027	0.9933 ± 0.0004	0.9947 ± 0.0006
2.3622	1.0021 ± 0.0031	1.0011 ± 0.0004	1.0000 ± 0.0006
2.6670	1.0028 ± 0.0031	0.9989 ± 0.0004	0.9975 ± 0.0006
2.9032	1.0033 ± 0.0046	1.0068 ± 0.0004	1.0057 ± 0.0006
3.3528	1.0041 ± 0.0055	1.0079 ± 0.0004	1.0061 ± 0.0005
3.5204	1.0043 ± 0.0052	1.0055 ± 0.0003	1.0032 ± 0.0005

Table V – Results of Sample Calculations, 16% ^{240}Pu

Pitch (cm)	Experimental k_{eff} with biases	KENO (27-Group ENDF/B-IV) k_{eff} ± Std. Deviation	MCNP (Continuous Energy ENDF/B-V) k_{eff} ± Std. Deviation
2.3622	1.0023 ± 0.0035	1.0023 ± 0.0003	1.0033 ± 0.0005
2.6670	1.0024 ± 0.0039	1.0006 ± 0.0003	0.9996 ± 0.0005
2.9032	1.0036 ± 0.0046	1.0035 ± 0.0003	1.0037 ± 0.0005
3.3528	1.0037 ± 0.0057	1.0039 ± 0.0003	1.0026 ± 0.0004
3.5204	1.0044 ± 0.0061	1.0025 ± 0.0003	1.0015 ± 0.0004

Table VI – Results of Sample Calculations, 24% ^{240}Pu

Pitch (cm)	Experimental k_{eff} with biases	KENO (27-Group ENDF/B-IV) k_{eff} ± Std. Deviation	MCNP (Continuous Energy ENDF/B-V) k_{eff} ± Std. Deviation
2.0320	0.9997 ± 0.0032	0.9937 ± 0.0004	0.9953 ± 0.0006
2.3622	1.0008 ± 0.0030	0.9984 ± 0.0004	0.9982 ± 0.0006
2.6670	1.0023 ± 0.0038	0.9998 ± 0.0004	1.0000 ± 0.0006
2.9032	1.0015 ± 0.0047	1.0041 ± 0.0004	1.0025 ± 0.0005
3.3528	1.0022 ± 0.0056	1.0059 ± 0.0004	1.0034 ± 0.0005
3.5204	1.0028 ± 0.0065	1.0051 ± 0.0004	1.0032 ± 0.0005

Tables VII and VIII give the difference between the calculated k_{eff} and the benchmark k_{eff} for each case with KENO-Va and MCNP. The standard deviation calculated for the difference between the calculated and benchmark k_{eff} is the square root of the sum of the squares of the benchmark uncertainty and the calculational uncertainty.

Table VII – Comparison of Benchmark and Calculated k_{eff}, KENO-Va

Pitch (cm)	8% ^{240}Pu Calculated k_{eff} - Benchmark k_{eff}	16% ^{240}Pu Calculated k_{eff} - Benchmark k_{eff}	24% ^{240}Pu Calculated k_{eff} - Benchmark k_{eff}
2.0320	-0.0081 ± 0.0027		-0.0060 ± 0.0032
2.3622	-0.0010 ± 0.0031	0.0000 ± 0.0035	-0.0024 ± 0.0030
2.6670	-0.0039 ± 0.0031	-0.0018 ± 0.0039	-0.0025 ± 0.0038
2.9032	0.0035 ± 0.0046	-0.0001 ± 0.0046	0.0026 ± 0.0047
3.3528	0.0038 ± 0.0055	0.0002 ± 0.0057	0.0037 ± 0.0056
3.5204	0.0012 ± 0.0052	-0.0019 ± 0.0061	0.0023 ± 0.0065

Table VIII – Comparison of Benchmark and Calculated k_{eff}, MCNP

Pitch (cm)	8% ^{240}Pu Calculated k_{eff} - Benchmark k_{eff}	16% ^{240}Pu Calculated k_{eff} - Benchmark k_{eff}	24% ^{240}Pu Calculated k_{eff} - Benchmark k_{eff}
2.0320	-0.0067 ± 0.0028		-0.0044 ± 0.0033
2.3622	-0.0021 ± 0.0031	0.0010 ± 0.0035	-0.0026 ± 0.0031
2.6670	-0.0053 ± 0.0031	-0.0028 ± 0.0039	-0.0023 ± 0.0038
2.9032	0.0024 ± 0.0046	0.0001 ± 0.0046	0.0010 ± 0.0047
3.3528	0.0020 ± 0.0055	-0.0011 ± 0.0057	0.0012 ± 0.0056
3.5204	-0.0011 ± 0.0052	-0.0029 ± 0.0061	0.0023 ± 0.0065

A plot of the difference between the calculated and benchmark k_{eff} values as a function of the lattice pitch is given in Figure 1 for KENO-Va and Figure 2 for MCNP. A general trend is shared for all three isotopic compositions and specifically by the 8% ^{240}Pu and 24% ^{240}Pu cases. At lower lattice spacings, calculated values are generally less than experimental, and at the 2.9032 and 3.3528 cm lattice spacings, the opposite is true. For the 3.5204 cm lattice spacing, the difference decreases again.

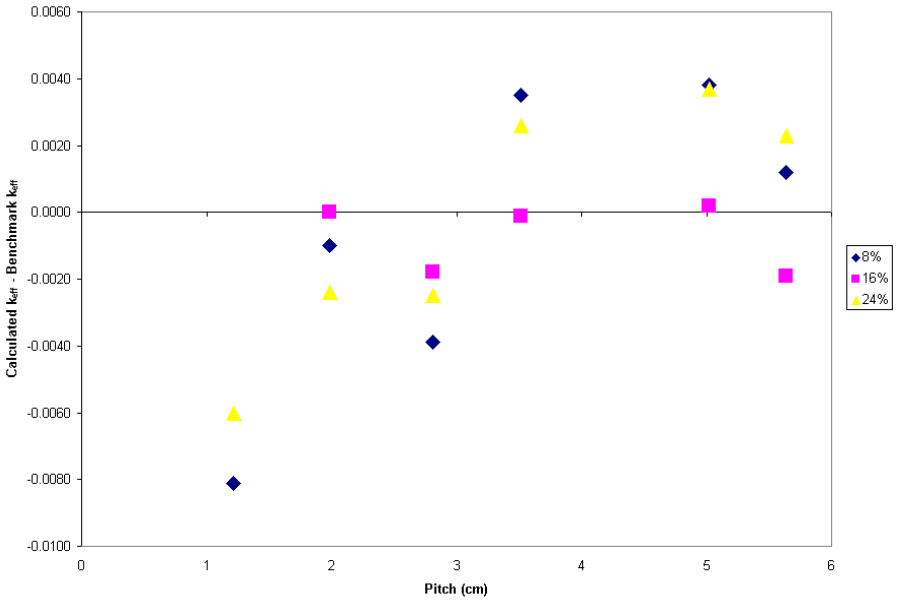

Figure 1 – Calculated k_{eff} – Benchmark k_{eff}, KENO-Va

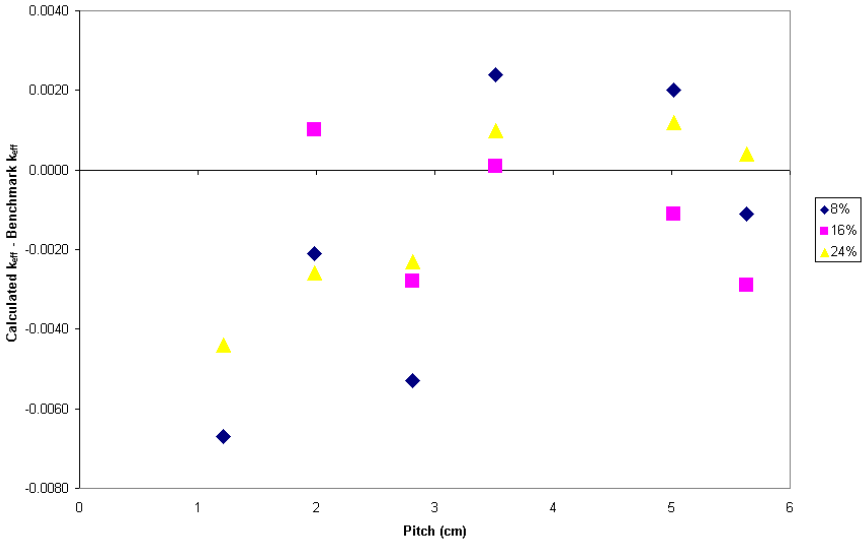

Figure 2 – Calculated keff – Benchmark keff, MCNP

The ^{239}Pu capture and fission cross sections have a large resonance (at 0.29 eV) just above the thermal Maxwellian peak of the flux spectrum in thermal reactors. These resonances can be major contributors to the reactivity effects due to changes in the fuel to moderator ratio (lattice pitch). A recent study[8] indicates that any calculation for fuel containing high amounts of plutonium and minor actinides should be considered with a high uncertainty margin. Finck[9] et al. compared the performance of ENDF/B-VI and JEF2.2 libraries for a series of MOX (mixed uranium/plutonium oxides) benchmarks for k value, void worth, and pin power distributions. They found that the differences in the ^{239}Pu cross sections between the two evaluations can lead to over 4 mk difference in reactivity for their benchmark configurations. We compared the integral cross section data from different evaluations (ENDF/B-VI, JEF2.2, JENDL-3.2, BROND-2 and CENDL-2) given in JEF Report 14[10], and observed as high as 3% difference in the resonance integral for the fission to capture ratio of ^{239}Pu. These observations lead one to believe that the uncertainty in the cross sections of this isotope is large enough to be a concern in terms of reactivity effects.

The existence of a shared trend in the difference between the calculated and benchmark k_{eff} as a function of pitch supports the observation above. That is, a bias from ^{239}Pu cross section uncertainties is being introduced into the calculated k_{eff}. This is also indicated by the fact that for the 8% ^{240}Pu and 24% ^{240}Pu cases the greatest difference between the calculated and benchmark k_{eff} is observed for the 2.0320 cm pitch lattices. A smaller pitch leads to a lower H/Pu ratio, which may shift the energy spectrum towards the 0.29 eV resonance peak of ^{239}Pu cross sections.

While a trend does appear in these results, the uncertainty in the difference between the calculated and benchmark k_{eff} is large. Therefore, the existence of the trend cannot be conclusively proven. A perturbation study of these cases is required to determine if any bias is being introduced into the calculated k_{eff} from uncertainties in cross sections, and to isolate the isotopes and energy ranges which cause such a bias if it exists.

Conclusions

Seventeen experiments involving mixed oxide fuel rods in water moderated triangular pitch lattices were evaluated as part of the ICSBEP. All seventeen experiments were acceptable as benchmarks for validation of cross sections and computer codes used in criticality safety. Benchmark models of the experiments were developed and the effect of uncertainty in the measured experimental data on k_{eff} was evaluated. Values of k_{eff} were found using these models with both MCNP with the ENDF/B-V continuous energy cross section library and KENO-Va with the 27-group ENDF/B-IV cross section library.

The difference between the results of these calculated values of k_{eff} and the benchmark values of k_{eff} follow a general trend as a function of the lattice pitch for the three values of ^{240}Pu isotopic composition studied. This indicates the possibility of the introduction of a bias in the calculated k_{eff} resulting from cross section uncertainties, although the magnitude of the uncertainty in this difference is large. A further study of these cases is

required to determine if the observed trend is caused by cross section uncertainties and to determine the cause of this trend.

References

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[b] The ICSBEP committee requires the use of the 14th edition rather than the 15th edition.