

Argonne National Laboratory

CONSISTENT HEAVY-ATOM CROSS-SECTION EVALUATION FOR PLUTONIUM RECYCLE CALCULATIONS

by

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CONSISTENT HEAVY-ATOM CROSS-SECTION EVALUATION FOR PLUTONIUM RECYCLE CALCULATIONS

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I. INTRODUCTION

The EBWR Plutonium Recycle Experiment has been initiated to provide data for the creation of a computational model that will permit accurate prediction of isotopic behavior and reactivity changes with burnup for plutonium-fueled, light-water, thermal reactors. A preliminary model, described in this report, has been tested against data from a number of cold critical experiments. Data from hot criticals and from the EBWR central plutonium region at various burnups are necessary before a complete model can be formulated.

The present model utilizes the GAM,⁽¹⁾ RIC,⁽²⁾ SOFOCATE,⁽³⁾ (or TEMPEST), DSN,⁽⁴⁾ and REX⁽⁵⁾ codes. This model has been tested against data both from the YANKEE critical experiments (with stainless-steel-clad UO₂ rods) and from Hanford Laboratories' critical experiments with zirconium-clad PuO₂-UO₂ rods and with plutonium-aluminum rods. Good agreement between the model and the experimental data has been obtained for the above-mentioned systems.

II. ANALYSIS OF YANKEE CRITICAL EXPERIMENTS^(6,7)

The YANKEE criticals consisted of six lattices with H₂O/UO₂ volume ratios varying from 1 to 5. The UO₂ fuel was arranged in pins of 0.381-cm radius, 122 cm in height, and clad with 0.0407-cm-thick stainless steel. The enrichment was 2.73 a/o U²³⁵. The four lattices with the lowest H₂O/UO₂ ratios were chosen for analysis.

Four energy groups were used in the calculation with break points at 5.53 keV, 1.44 eV, and 0.532 eV. These break points were chosen as appropriate for plutonium systems containing a considerable fraction of Pu²⁴⁰. The normal GAM library⁽⁸⁾ was employed (P-1 option) to obtain cross sections for the upper three groups except that resonance parameters were included in the library for Pu²⁴⁰ (material No. 132) and U²³⁵ (material No. 133), and the Hanford RBU Library⁽⁹⁾ values were employed for U²³⁸ (material No. 139). The resonance integral of Pu²⁴⁰ as a function of Pu²⁴⁰ concentration has been compared with Nichols' experimental data⁽¹⁰⁾

and found to be in agreement. GAM's U^{238} (material No. 12) has a deficiency in the capture cross section in the unresolved resonance region (1 keV \rightarrow 100 keV), which gives rise to an overestimation of the reactivity for each lattice (see Fig. 1).

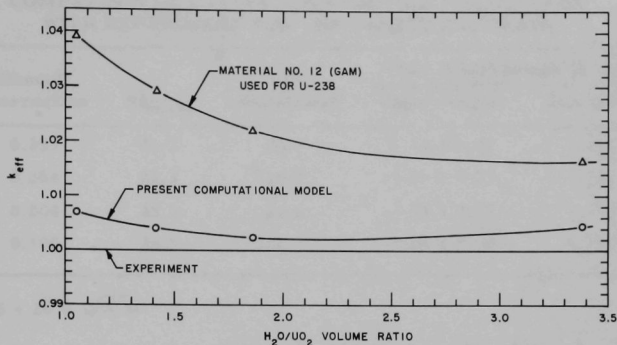


Fig. 1. Comparison of Computational Model with YANKEE Critical Experiments

Initial thermal-group cross sections were obtained from a preliminary SOFOCATE which used the actual isotopic densities as input. These cross sections were employed to prepare input for an S_8 DSN problem that provided flux weighting factors for the various reactor materials. An extra pure-scattering region, four mean-free-paths thick, surrounded the actual DSN cell. A final SOFOCATE, which utilized flux-weighted isotopic densities as input, provided the thermal-group cross sections. The hydrogen gas moderator option (301) was used. The U^{235} library cross sections were obtained from BNL-325 (Supplement 1) with ν^{25} such that η^{25} (2200 m/sec) was brought into agreement with the World Consistent Set value of 2.07.

The input buckling (B_g^2) to both GAM and SOFOCATE was the critical buckling for the lattice as obtained from the experiment. A one-dimensional, diffusion-theory code (REX) was used to obtain the multiplication factor at the experimental critical radius. For two of the lattices, cross sections were also obtained using only the vertical buckling as input. For each case, the resultant calculated reactivity changed by less than 0.1%. The reactivity predicted by the calculational model is compared to experiment in Fig. 1.

The calculated Dancoff correction and the resonance integral from GAM for U^{238} are given in Table I. Hellstrand's RI value (corrected by the same Dancoff factor) is also listed for each lattice. The Dancoff factors were calculated neglecting the cladding of the fuel. Incorporation of the cladding effect results in a somewhat higher value for C , and thus an

increase in k_{eff} (calculated). This effect is nonnegligible for tight lattices (+0.5% for $\text{H}_2\text{O}/\text{UO}_2 = 1.05$) but much smaller at higher volume ratios.

Table I

COMPARISON OF CALCULATED LATTICE PARAMETERS
WITH EXPERIMENT FOR THE YANKEE CRITICALS

$\text{H}_2\text{O}/\text{UO}_2$	Dancoff Correction	RI_{GAM}	RI^* (Hellstrand)	Fuel Disadvantage Factor		$\overline{\eta f}$
				Experimental	Calculated	
1.05	0.333	21.2	20.0	1.14 ± 0.03	1.118	1.515
1.41	0.264	22.3	20.7	1.16 ± 0.03	1.129	1.494
1.86	0.204	23.1	21.4	1.17 ± 0.03	1.140	1.466
3.36	0.100	24.5	22.7	1.25 ± 0.08	1.170	1.374

$$*\text{RI} = 4.15 + 26.6 \sqrt{S/M}$$

The fuel disadvantage factor is experimentally defined as⁽⁷⁾

$$\text{DF}_{\text{exp}} = \frac{V_F \int_{V_M} \int_0^{0.5} \sigma_F^{25}(E) \phi(E, V) dE dV}{V_M \int_{V_F} \int_0^{0.5} \sigma_F^{25}(E) \phi(E, V) dE dV}$$

This is compared to the value obtained from DSN for each lattice in Table I; the calculated values of $\overline{\eta f}$ are also given. The experimental disadvantage factor is expected to be larger than the calculated result since the former is based on foil activations. Better agreement would be expected if the calculations used the THERMOS code.⁽¹¹⁾

III. ANALYSIS OF HANFORD'S CRITICAL EXPERIMENTS WITH PuO_2 - UO_2 FUEL ELEMENTS⁽¹²⁾

The Hanford Laboratories' criticals with PuO_2 - UO_2 fuel were performed with fuel pins almost identical to those that will be used in the EBWR Plutonium Recycle Experiment. The fuel was arranged on a triangular pattern using pins of 0.472-cm radius, 123 cm in height, and clad with 0.0685-cm-thick zirconium. The isotopic enrichments were: 1.366 a/o Pu^{239} , 0.117 a/o Pu^{240} , 0.011 a/o Pu^{241} , 0.219 a/o U^{235} , and 98.287 a/o U^{238} . Four lattices were examined with $\text{H}_2\text{O}/\text{UO}_2$ volume ratios varying from 1 to 5.

The energy group structure and the calculational model were the same as for the analysis of the YANKEE criticals. The Illinois Institute of Technology Research Institute resonance integral code (RIC) was employed

to obtain self-shielding factors as a function of lattice pitch for Pu^{239} in group 2. This code evaluates Doppler-broadened resonance integrals in a manner similar to that used in GAM. The self-shielding factors were used as input for GAM. The vertical buckling was chosen as the input buckling for GAM and SOFOCATE. The Dancoff correction was calculated utilizing an effective scattering cross section (for the moderator and clad mixture) which is dependent upon the relative thicknesses of H_2O and zirconium between pin centers. Calculated lattice parameters are listed in Table II.

Table II

CALCULATED LATTICE PARAMETERS FOR
HANFORD'S PuO_2 - UO_2 CRITICALS

$\text{H}_2\text{O}/\text{UO}_2$	Dancoff Correction	$\text{RI}_{\text{GAM}}^{238}$	$\text{RI}_{\text{Hellstrand}}^{238}$	$\text{RI}_{\text{GAM}}^{240}$	$\overline{\eta f}$
1.1	0.326	19.6	18.8	4390	1.605
2.7	0.113	22.6	21.0	4920	1.488
3.8	0.065	23.2	21.4	5020	1.409
5.1	0.037	23.5	21.7	5080	1.317

The thermal cross-section data for Pu^{240} and Pu^{241} were taken from BNL-325, while the data for Pu^{239} were adjusted to bring the calculation into agreement with the experimental multiplication factor for the largest $\text{H}_2\text{O}/\text{UO}_2$ volume ratio. From the data in BNL-325 (Supplement 1), a so-called α^{49} ("BNL Best") was chosen as a function of energy. The "best" value for any energy was taken to be an average between (1) the values obtained using the $\sigma_{\text{abs}}^{49} \sqrt{E}$ and $\sigma_{\text{F}}^{49} \sqrt{E}$ curves, and (2) an average of the direct experimental points. The α values at each energy were then multiplied by an appropriate factor which would bring the calculational model into agreement with experiment for the lattice with the $\text{H}_2\text{O}/\text{UO}_2$ volume ratio equal to 5.14. The recommended α^{49} values as a function of energy are shown in Fig. 2. The use of these α^{49} values, the BNL-325 σ_{f}^{49} values, and $\nu^{49} = 2.89$ gives good agreement with experiment, as is shown in Fig. 3.

In Table III, various 2200-m/sec cross-section sets for Pu^{239} are compared. Sher's set has been found by Hanford Laboratories to be in good agreement with experiment.⁽¹²⁾ Aline's set was that recommended by him after a study of Hanford's 1.82 w/o plutonium-aluminum subcritical experiments.⁽¹³⁾ Note that the cross-section set obtained by the previously described normalization is in good agreement with the World Consistent Set.

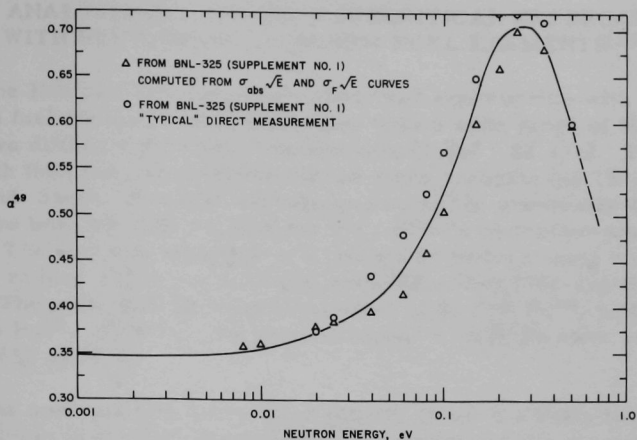


Fig. 2. α^{49} versus Neutron Energy

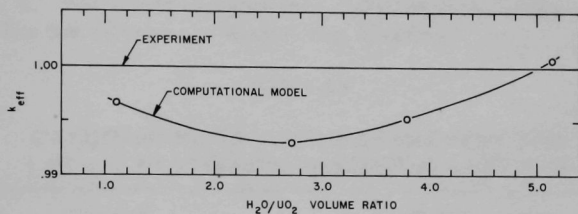


Fig. 3. Comparison of Computational Model with Hanford Critical Experiments Using $\text{PuO}_2\text{-UO}_2$ in H_2O

Table III

2200-m/sec CROSS-SECTION DATA FOR Pu^{239}

	Aline (Set II)	Sher	World Consistent	Moon
σ_a	1008	1030	1028 ± 8	1023
σ_f	726	748	742 ± 4	740
ν	2.89	2.88	2.89 ± 0.03	2.89
η	2.082	2.092	2.086	2.09
α	0.3884	0.3770	0.3854	0.383

IV. ANALYSIS OF HANFORD'S SUBCRITICAL EXPERIMENTS WITH PLUTONIUM-ALUMINUM FUEL ELEMENTS⁽¹⁴⁾

The Hanford Laboratories' subcritical experiments with plutonium-aluminum fuel elements were performed over a wide range of H/Pu ratios and for two different plutonium enrichments (5 and 1.82 w/o). The experiments with the lower enrichment are the more accurate ($\pm 0.1\%$ uncertainty in k_{eff}) and, hence, extensive calculations using the previously described model have been carried out only for the 1.82 w/o plutonium-aluminum lattices. The fuel was arranged in a triangular pattern using pins of 0.635-cm radius, 111.3 cm in height, and clad with 0.0762-cm-thick zirconium. The plutonium by weight consisted of 93.97% Pu²³⁹, 5.58% Pu²⁴⁰, and 0.44% Pu²⁴¹. Five lattices were examined with H/Pu atom ratios varying from 630 to 1418.

The analysis was carried out exactly as for the PuO₂-UO₂ lattices. The aluminum in the rod was considered a moderator for the purpose of evaluating the resonance absorption in Pu²³⁹ and Pu²⁴⁰. Table IV lists various lattice parameters. The calculational results are compared to experiment in Fig. 4. The error is relatively constant with lattice spacing but larger than for the previously considered systems.

Table IV

CALCULATED LATTICE PARAMETERS FOR
1.82 w/o PLUTONIUM-ALUMINUM LATTICES

H/Pu	Dancoff Correction	RI ²⁴⁰	Disadvantage Factor of Fuel	$\bar{\eta}f$
632.5	0.217	6490	1.091	1.506
812.8	0.158	6580	1.095	1.445
1004.5	0.117	6630	1.099	1.384
1208.1	0.087	6670	1.104	1.323
1423.3	0.066	6690	1.108	1.264

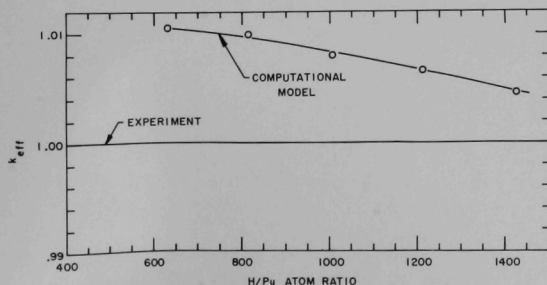


Fig. 4

Comparison of Computational
Model with Hanford Subcritical
Experiments Using 1.82 w/o Pu-Al
in H₂O

V. CONCLUSIONS

The calculational model described herein is in good agreement with experiment for cold, clean $\text{PuO}_2\text{-UO}_2$ systems over a wide range of $\text{H}_2\text{O}/\text{UO}_2$ volume ratios. It seems likely that there are certain small compensating errors. The resonance integral of U^{238} has probably been overestimated, while the disadvantage factor of the fuel has probably been underestimated. Since there is no U^{238} in the plutonium-aluminum lattices, such an underestimation might well have led to the slight overestimation of k_{eff} shown in Fig. 4. More accurate thermal calculations and evaluation of the U^{238} resonance integral for clad rods would help to increase the accuracy of the model.

The η^{49} and α^{49} values given in this report are in good agreement with experiment and are consistent with the World Consistent Set. The shape of α^{49} as a function of energy may well be modified slightly as data become available from hot criticals.

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