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PuO_2 GRAIN REACTIVITY EFFECTS

F. R. CZERNIEJEWSKI

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PuO_2 GRAIN REACTIVITY EFFECTS

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June 1967

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PuO₂ GRAIN REACTIVITY EFFECTS

F. R. Czerniejewski

INTRODUCTION

A series of experiments will be carried out by Battelle-Northwest in the Physical Constants Test Reactor (PCTR) to determine the reactivity effects of the size of PuO₂ grains imbedded in a natural UO₂ rod in a graphite moderator. The PCTR experiments will help answer several questions in the Plutonium Utilization Program concerning the importance of PuO₂ grain size in future fuel fabrication specifications. A theoretical study was initiated with the experimental program to predict the reactivity effects of fuel particle size.

SUMMARY

The theoretical study presented here gives a model for predicting k_{∞} from basic cross-section data and from the size and composition of the experimental cell. Resonance self-shielding for individual particles is calculated from collision probabilities, and the balance equation is solved by Simpson's integration. When the terms of the balance equation are suitably arranged, iteration is necessary only in the thermal energy range. Presently, this method has not been verified. However, since most of the usual simplifying assumptions have been removed, the results should be in reasonable agreement with measurements. If verified, the calculational method presented in this report should be of value in predicting PuO₂ grain effects beyond the range of the experiments.

THEORY

Define the collision rate in the i^{th} material (UO₂, PuO₂, or C) as

$$F_i(E) = V_i \sum_i^t (E) \phi_i(E) \quad (1)$$

Then, as usual in slowing down calculations, one may write⁽¹⁾

$$F_i(E) = \sum_j^{\text{materials}} P_{j \rightarrow i}(E) \sum_{k/j}^{\text{elements}} \int_E^{E/\alpha_k} \frac{F_j(E') p_{k/j}(E') dE'}{(1-\alpha_k) E'} \quad (2)$$

for neutrons undergoing isotropic scattering above thermal energies. In Equation (2),

$P_{j \rightarrow i}(E)$ = probability that a neutron born at E in material j makes its next collision in material i ,

$$\alpha_k = \left(\frac{A_k - 1}{A_k + 1} \right)^2$$

where A_k = ratio of atomic mass of k^{th} element to neutron mass,

k/j indicates the k^{th} element in the j^{th} material, e.g., O_2/UO_2 , and

$$p_{k/j}(E') = \frac{\sum_k^s(E')}{\sum_j^t(E')} \quad ,$$

is the ratio of the scattering cross section of the k^{th} element to the total cross section of material j . Changing to lethargy and approximating the integral in Equation (2) by a numerical sum (Simpson's rule, for example), we can derive a simultaneous set of equations at each lethargy, u_j .

Define

$$Z_{ii}(u_j) = \frac{\Delta u}{3} \sum_{k/i}^{\text{elements}} \left[\frac{p_{k/i}(u_j)}{1 - \alpha_k} \right] \quad (3)$$

and let

$$\begin{aligned}
 Q_1(u_j) &= \frac{\Delta u}{3} \left[\frac{I_{U/UO_2}(u_j)}{1 - \alpha_U} + \frac{I_{O_2/UO_2}(u_j)}{1 - \alpha_O} \right] \\
 Q_2(u_j) &= \frac{\Delta u}{3} \left[\frac{I_{Pu/PuO_2}(u_j)}{1 - \alpha_{Pu}} + \frac{I_{O_2/PuO_2}(u_j)}{1 - \alpha_O} \right] \\
 Q_3(u_j) &= \frac{\Delta u}{3} \frac{I_C(u_j)}{1 - \alpha_C}
 \end{aligned} \tag{4}$$

where Simpson's rule approximates the integral in

$$I_{k/i}(u_j) = \int_{u_j - d_k}^{u_j} F_i(u') p_{k/i}(u') du' - Z_{ii}(u_j)$$

and $d_k = -\ln \alpha_k$ is the maximum lethargy change for scatter from element k .

If Δu is chosen such that $\frac{-\ln \alpha_C}{\Delta u}$ is an integer, the maximum lethargy increase for scatter from U, Pu, and O is not in general an integral number of Δu 's. In that case, the quadratic fit used for Simpson's rule may be integrated for the noninteger limits on one end of the slowing down range by the following general formula:

$$\begin{aligned}
 \int_{ah}^{2h} T(u') du' &\approx \frac{h}{3} \left\{ T(0) \left[1 + \frac{9}{4}a^2 - 3a - \frac{1}{2}a^3 \right] \right. \\
 &\quad \left. + T(h) \left[4 + a^3 - 3a^2 \right] + T(2h) \left[1 - \frac{1}{2}a^3 + \frac{3}{4}a^2 \right] \right\} \\
 &\quad \text{for } 0 \leq a \leq 2.
 \end{aligned}$$

(5)

We then have for each lethargy the simultaneous set

$$(\underline{I} - \underline{P}\underline{Z})\underline{F} = \underline{P}\underline{Q}, \quad (6)$$

where

$\underline{I}$ is the unit diagonal matrix

$\underline{P}$ is the region to region transfer matrix with $P_{j \rightarrow i} = P_{ij}$ in the matrix

$\underline{Z}$ is the diagonal matrix defined by Equation (3)

$\underline{Q}$ is a column vector

$\underline{F}$ is the unknown column vector of collision rates.

By the placement of the term $\underline{P}\underline{Z}$ on the left, Equation (6) is formulated so that the j^{th} component of $\underline{F}$ may be determined uniquely in terms of the first to $(j-1)^{\text{th}}$ components, and iteration is not required in the slowing down region. The calculation assumes a $1/E$ flux at some reasonably high energy (this implies no absorption) and then proceeds from one energy to the next lower energy by solution of Equation (6). Since small effects are involved, the number of points chosen per maximum energy change in graphite was 1200, giving $\Delta u \approx 3 \times 10^{-4}$. The energy for beginning the non $1/E$ calculation is about 175 eV, hence nearly 20,000 energies are used. The effect of particle size comes into the calculation via the $P_{i \rightarrow j}$ matrix. This matrix is calculated following Lewis and Connolly.⁽³⁾ The method of calculation is reproduced in the Appendix

Inasmuch as spatial flux dependence is ignored in the calculation of $P_{i \rightarrow j}$, the set $F_i(E)$ is used to generate flux-averaged broad group cross sections for a transport or diffusion code.

In the thermal range, the theory and solution method remain the same as above except for two changes. The slowing down kernel $\sum_s (E')/(1 - \alpha)E'$ is replaced by a thermal kernel $\sum_s (E' \rightarrow E)$ which allows for up scatter and calculated theoretically. Iteration⁽⁴⁾ on the set of $F_i(E)$ must be performed since the solution of Equation (6) at one energy no longer

leads to the equation for the next lower energy. However, convergence is rapid if a Maxwellian is initially assumed. Sources are found by proper integration of the slowing down of the epithermal neutrons. Thermal cross sections for a homogeneous rod are then obtained by averaging as above.

The computer code GRAINS has been written in Fortran IV for the UNIVAC 1107 to solve Equation (6) for the collision densities and to average the cross sections.

RESULTS

Figure 1 shows the dimensions of the physical system measured in the PCTR. Figure 2 shows calculated curves of k_{∞} versus particle size from diffusion theory (HFN) and the flat flux approximation.

FIGURE 1. Dimensions of Physical System in PCTR

FIGURE 2. k_{∞} Versus Particle Size; 2% PuO_2 Particles in Natural UO_2 1/2 in. Rod, 4 in. Graphite Lattice

For these curves, a single fast group, 10 MeV - 167 eV, was calculated by HRG⁽⁸⁾ using a homogeneous rod model. Four epithermal groups and a thermal group were obtained from GRAINS. Densities used for UO_2 and PuO_2 were 80% of theoretical.

GRAINS CODE

GRAINS is operative with the following input with the tape assignments. Note that * items are not variable unless the code, and/or data tapes are altered.

<u>Card</u>	<u>Format</u>	<u>Meaning</u>	<u>Typical Input</u>
1	I 10	M, number of energy points in epithermal range	20401*
1	I 10	JP, number of maximum energy transfers in epithermal range	17*
1	I 10	NGPS, number of epithermal broad groups	4*
2	E 10.6	EBOT, thermal cutoff (eV)	0.683*
2	E 10.6	RROD, rod radius (cm)	0.635
2	E 10.6	PCELL, cylindricized cell radius (cm)	5.732
2	E 10.6	PCTU, fraction ^{235}U in U	0.0072
2	E 10.6	WTN, neutron atomic weight	1.0086654*
2	E 10.6	VA, volume fraction of particle in rod	0.02
2	E 10.6	WT(I), atomic weight of i^{th} isotope	
3	E 10.6	SIGP(I), potential scattering microscopic cross section for i^{th} isotope (barns)	
3	E 10.6	FIS(I), Nu for i^{th} isotope	
Repeat Card 3 for 6 isotopes in order: ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{16}O , ^{12}C			
4	3 E 10.6	DENS(J), J = 1,3. Densities of UO_2 , PuO_2 , C in order (gm/cc)	
5	5 E 10.6	ET(K), K = 1,5. Highest thermal energies	0.683, 0.63, 0.57, 0.532, 0.49*

<u>Card</u>	<u>Format</u>	<u>Meaning</u>	<u>Typical Input</u>
6	I 5	LAST, 1 if last case, 0 if not	
6	I 5	NOENGY, number of thermal energies	57*
6	I 5	LIMIT, maximum iterations for thermal calculation	100
6	I 5	LTAPE, if 1 put integrals used for cross section averaging on tape	0
6	E 10.6	RSPH, radius of spherical particles (cm)	0.035
6	E 10.6	CONV, convergence criteria for thermal iteration	0.0001
6	E 10.6	PCTPU, fraction of ^{240}Pu in Pu	0.08

Only Card 6 needs be repeated for additional cases.

Each case runs about 18 minutes.

GRAINS OUTPUT

GRAINS lists the average macroscopic cross sections for the rod and moderator (except those considered constant in the moderator) for the epithermal groups specified by NGPS and a thermal group. These cross sections, together with a fast (above 160 eV) group constitutes the input to a transport or diffusion code. GRAINS also prints out an approximate value of k_{∞} based on: 1) flat spacial fluxes, 2) the epithermal-thermal energy range only.

SUGGESTED IMPROVEMENTS

Possible improvements in the GRAINS code, or a successor to it, follow:

- (1) Generalization of the system to accept water moderator, particles in the moderator, changes in temperature, and anisotropic flux.
- (2) Calculation of cross sections for each isotope.
GRAINS first smears the U and Pu cross sections by relative amounts of each isotope.

- (3) Possible direct accounting for space dependence either by changing $P_{i \rightarrow j}$ or by subdividing⁽²⁾ regions and calculating the collision rate $F(E)$ for each subdivision. For the natural uranium rod to be used in the PCTR experiment, the thermal flux depression factor is only about 1.2 thus, the spatial effect is not too important.

APPENDIX

CALCULATION OF COLLISION PROBABILITIES

The calculation of collision probabilities follows Lewis and Connolly.⁽³⁾ The nine collision probabilities, $P_{i \rightarrow j}$ for $i = 1, 2, 3$, and $j = 1, 2, 3$, are calculated assuming a flat source density in each region.

Under this assumption, the first-flight uniform-isotopic source escape probabilities for a sphere and a cylinder from Case⁽⁵⁾ may be used:

$$P_s = \frac{3}{8x} [2x^2 - 1 + (1 + 2x)e^{-2x}]$$

$$P_c = \frac{2x}{3} \left(2 \{x[K_1(x)I_1(x) + K_0(x)I_0(x)] - 1\} + \frac{1}{x}K_1(x)I_1(x) - K_0(x)I_1(x) + K_1(x)I_0(x) \right),$$

where x = radius times average total cross section, and the K and I are Bessel functions. Furthermore, the reciprocity relation⁽⁶⁾ states

$$P_{i \rightarrow j} \sum_i^t V_i = P_{j \rightarrow i} \sum_j^t V_j$$

and, of course,

$$\sum_{j=1}^3 P_{i \rightarrow j} = 1 \quad i = 1, 2, 3.$$

The PuO_2 to UO_2 transfer probability is expressed as

$$P_{2 \rightarrow 1} = P_s(1 - P_c)P_{1/r}$$

where $P_{1/r}$ is the conditional probability that a neutron colliding in the rod collides with UO_2 . This probability is

$$P_{1/r} = \frac{\Sigma_1 V_1}{\Sigma_1 V_1 + P_s \Sigma_2 V_2},$$

and

$$P_{2 \rightarrow 3} = P_s P_c$$

so

$$P_{2 \rightarrow 2} = 1 - P_{2 \rightarrow 1} - P_{2 \rightarrow 3} .$$

The probability for escape from the UO_2 into the C is

$$P_{1 \rightarrow 3} = P_c .$$

By the reciprocal relation

$$P_{1 \rightarrow 2} \sum_1^t V_1 = P_{2 \rightarrow 1} \sum_2^t V_2 ,$$

$$P_{1 \rightarrow 2} = P_{2 \rightarrow 1} \frac{\sum_2 V_2}{\sum_1 V_1} .$$

Hence,

$$P_{1 \rightarrow 1} = 1 - P_{1 \rightarrow 2} - P_{1 \rightarrow 3} .$$

The remaining probabilities are found from the reciprocal relation

$$P_{3 \rightarrow 1} = P_{1 \rightarrow 3} \frac{\sum_1 V_1}{\sum_3 V_3}$$

and

$$P_{3 \rightarrow 2} = \frac{P_{2 \rightarrow 3} \sum_2 V_2}{\sum_3 V_3} .$$

Thus,

$$P_{3 \rightarrow 3} = 1 - P_{3 \rightarrow 1} - P_{3 \rightarrow 2} .$$

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