CRITICAL PARAMETERS OF PLUTONIUM SYSTEMS. PART I: ANALYSIS OF EXPERIMENTS

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To predict the critical parameters of plutonium fueled systems one must establish the accuracy of the computational methods to be employed and the accuracy and applicability of the available critical experiment data with which the calculations are to be compared. The accuracy of a multigroup diffusion theory code, HFN, and a multigroup transport theory code, DTF-IV, was examined by analyzing recent plutonium critical experiments. The experiments cover the entire range of possible moderation ratios, and the plutonium fuels contain as much as 23.2 isotopic percent ^{240}Pu. All three basic geometries are represented by the experimental data examined. Where necessary, the criticality data were corrected, by means of additional experiments and/or calculations, to conform to one-dimensional, clean, homogeneous critical assemblies which could be adequately defined and used as a basis for establishing nuclear criticality safety guidelines.

INTRODUCTION

A number of clean critical experiments have been performed in recent years on homogeneous, plutonium-fueled systems. These experiments indicate the need for revising much of the data that have been used as a guide for establishing nuclear criticality safety specifications.[1] In particular, recent experimental data show that the values given for critical dimensions and masses are quite conservative, especially for systems in the intermediate neutron energy range ($1 < H/Pu < 250$). These more recent data likewise can provide a basis for extending the existing criticality predictions to higher ^{240}Pu contents and to other moderation ratios.

In order to correct, and extend, the existing widely used criticality data, theoretical calculations are required to predict the critical parameters of systems for which no experimental data are available. The reliability of these predictions must be determined from the consistency with which the computational methods reproduce the widest possible variety of experimental criticality data. The purpose of this paper is to analyze some of the calculational techniques in use at our critical mass laboratory by comparing computed critical parameters with values measured, or derived, for clean, homogeneous, plutonium-fueled systems.

COMPUTATIONAL METHODS AND CROSS SECTIONS

Two one-dimensional multigroup computer codes were employed in analyzing the available experimental data. These were HFN, a diffusion theory code,[2] and the transport theory code[3] DTF-IV. In calculations with the latter code, the transport equation was solved in the S_4 approximation with anisotropic scattering defined through the first Legendre moment of the scattering kernel. Both codes utilized multigroup constants (18 energy groups) obtained from the GAMTEC-II code.[4] The HFN calculations were made using 50 spatial mesh points in the core region with an additional 20 mesh points, when applicable, in the reflector region. The DTF-IV code used 50 spatial mesh points total for all systems. For reflected systems, 30 mesh points were specified in the core region and 20 points were included in the reflector region. Both codes were converged in all cases to within 0.1 mk.

Constants for the 17 fast groups ($E > 0.683$ eV) were averaged over a 66-group slowing down spectrum computed using the B_1 approximation to the Boltzmann equation. The source was defined by the ^{239}Pu fission spectrum. Resonance absorption by ^{240}Pu was computed using resonance parameters obtained from the GAM-I library using both the narrow resonance and narrow resonance infinite mass approximations.

One thermal group was assumed ($E < 0.683$ eV), for which group constants were averaged over a Wigner-Wilkins spectrum. A preliminary investigation indicated that the most accurate results would be obtained using thermal ^{239}Pu cross-section data normalized to the 0.0253 eV values obtained from a 1962 least-squares analysis by Sher.[5]

This investigation consisted of comparing the effective multiplication factors computed using various thermal and epithermal ^{239}Pu cross-section data for two measured critical systems: PuO_2-polystyrene[6] (2.2 wt% ^{240}Pu, H/Pu = 15) and a spherical $Pu(NO_3)_4$ solution system[7] (4.6 wt% ^{240}Pu, 32.2 g Pu/liter). The additional data considered in this survey were those reported by Westcott,[8] Drake and Dyos,[9] Leonard,[10] and Sher[11] (the last in 1965). The effective multiplication factors computed using these data were found to be in error by 1 to 5.4% more than were the values obtained using the data which were normalized to the Sher 1962 values.

CORRELATION OF THEORY AND EXPERIMENT

Theoretical calculations are required to predict the critical parameters of systems for which no experimental data are available. The results of these calculations will be reliable only if a wide variety of experimental data can be reproduced with consistency. Since computational methods are applicable primarily to idealized systems, and since criticality safety specifications are based primarily on idealized systems, it is necessary to correct the experimental data so that the cleanest possible systems are represented. The theoretical results can be compared with these corrected experimental data to provide an unambiguous judgement on the validity of the computational techniques. If the result of these comparisons is favorable, the calculations may be used to provide guidance in establishing nuclear criticality safety specifications.

Well Moderated Systems

It can be seen from an examination of the results obtained by Richey[12] and by Lloyd et al.[7] that the HFN diffusion theory code will reproduce to a good degree of accuracy the experimentally determined critical parameters of spherical, well moderated $Pu(NO_3)_4$ solutions with ^{240}Pu contents of ≤ 5 wt%. Indeed, the mean value of k_{eff} obtained from the multitude of spherical systems examined was 1.000 ± 0.004.[12]

It should be noted that Richey[12] also examined the accuracy of the DTF code for these same systems. These calculations, however, did not reproduce the experimental data as well as did the diffusion theory calculations. The effects of using higher order S_N approximations and a higher order allowance for the anisotropic scattering by hydrogen were examined as a possible cause of the errors associated with the DTF calculations. It was concluded that for well moderated spheres the DTF calculations were adequately converged (within 3 mk) using the S_4 approximation with anisotropic scattering defined through the P_1 approximation.

To determine if critical parameters can be accurately predicted for semi-infinite cylinders and for higher ^{240}Pu contents in well-moderated systems, values of k_{eff} were computed, using the HFN code, for the $Pu(NO_3)_4$ solution critical experiments reported by Smith.[13] These solutions contained 13.51 wt% ^{240}Pu and had plutonium concentrations ranging ≤ 216 g Pu/liter. The cylinders were unreflected, with diameters of 13.4 and 17.9 in. Computed values of k_{eff} ranged from 0.986 to 1.002, and were examined as a function of the axial buckling of the cylinder.

This evaluation of the data is based on the assumption that the DB^2 leakage correction, necessary in the one-dimensional HFN code, introduces an error in the calculations. It would seem appropriate, therefore, to examine the variation of k_{eff} not only as a function of the magnitude of the DB^2 correction, but also as a function of the fraction of the total leakage which this correction represents.

Figure 1 shows the dependence of computed multiplication factors on the ratio of axial to total buckling for both these systems and those reported by Bruna[14] (1.5 wt% ^{240}Pu), which have been analyzed by Richey.[12] The assumed linear nature of the variation is most valid for small values of the ratio and departs from linearity as the ratio increases. If one uses only the data for which the linear relationship is apparent ($B_A^2/B_M^2 \leq 0.4$, see Fig. 1), then extrapolation, by means of a least-squares fit, to $B_A^2 = 0.0$ yields a k_{eff} of 1.0041 for the 13.51 wt% ^{240}Pu data, and 1.0044 for the 1.5 wt% ^{240}Pu data. Each data point shown in Fig. 1 represents a different $Pu(NO_3)_4$ solution critical experiment.

The values of k_{eff} that would be computed for

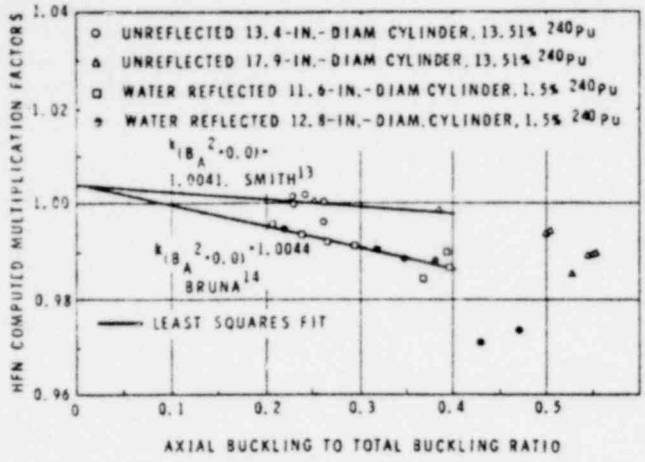

Fig. 1. Comparison of computed multiplication factors for cylindrical systems.

infinite length cylinders ($B_A^2 = 0.0$) are in good agreement despite the large difference in ^{240}Pu concentration. They also agree well with the average k_{eff} (1.00 ± 0.004) computed for the 4.6 wt% ^{240}Pu, spherical, $Pu(NO_3)_4$ solution data. It appears, therefore, that the critical parameters of well moderated, plutonium fueled systems can be accurately predicted in spherical and semi-infinite cylindrical geometries for systems containing up to ~20 wt% ^{240}Pu.

Critical parameters of semi-infinite slabs have been estimated by extrapolation from critical experiments performed in slab geometry using $Pu(NO_3)_4$ solutions containing 4.6 and 23.2 wt% ^{240}Pu.[15,16] For the reported, clean, water reflected slabs of $Pu(NO_3)_4$ solution, the HFN code computed effective multiplication factors which were high by as much as 2.7%. The multiplication factors computed for the reported, clean, unreflected slabs were low by ~7%. These results indicate that the effects of the stainless steel which contains and supports the $Pu(NO_3)_4$ solution, and the effects of room return neutrons, have been either experimentally or analytically underestimated.

An experimental determination of the effects of the steel tank walls and support structure has been previously reported.[17] These results were used to correct the measured, critical, infinite slab thicknesses to the clean, critical, infinite slab thicknesses reported by Lloyd et al.[15,16]

The effect of the support structure was determined by adding steel adjacent to the steel contained in the support structure [see Fig. 2(a)]. Since the addition of stainless steel in this manner does not significantly increase the probability that a neutron that escapes from the solution will impinge on the stainless steel, the effect of the added thickness of steel is greatly underestimated. Also, particularly in the case of the reflected systems, the additional thickness of steel increases the flux depression in the steel and thereby reduces the effect such that it is not representative of the thickness added.

Experiments to re-evaluate the effect of materials extraneous to these $Pu(NO_3)_4$ solution systems were recently performed with another $Pu(NO_3)_4$ solution containing ~241 g Pu/liter (18.4 wt% ^{240}Pu) with a free acid molarity of ~4.14. These experiments utilized a water reflected slab that was 5.80 in. thick and an unreflected slab 8.35 in. thick. When stainless-steel plates were added, they were placed diagonally across the square matrix of the support structure as shown in Fig. 2(b). The effect of these additional plates should be more representative of the effect of the steel contained in the support structure matrix. For the reflected system, the additional steel increased the critical height by 4.419 ± 0.035 in. The critical height of the unreflected system decreased by 6.059 ± 0.032 in. These changes in critical height are equivalent to changing the critical thicknesses by +0.115 ±0.005 in. and -0.230 ±0.005 in. for the reflected and unreflected systems, respectively (see Fig. 3).

An attempt was also made to compute the effect of the additional steel by uniformly distributing the stainless steel, on a volume basis, through the region enclosed by the support structure. The steel support structure occupied 5.90% of the total volume contained in the 4.5-in.-thick region adjacent to the slab walls. Upon insertion of the additional steel, 9.75% of this volume was occupied. The change in critical thickness due to this ~65% increase in steel was computed to be 0.104 in. for the reflected case, which compares favorably to the measured change of 0.115 in. For the unreflected system, the change was computed to be 0.350 in. as compared to the measured change of 0.230 in. This large error in the computed effect for the unreflected system arises from the fact that this system can not be represented as a diffusing medium, thereby introducing a large error into the multigroup diffusion theory calculations. The effect of the support structure for

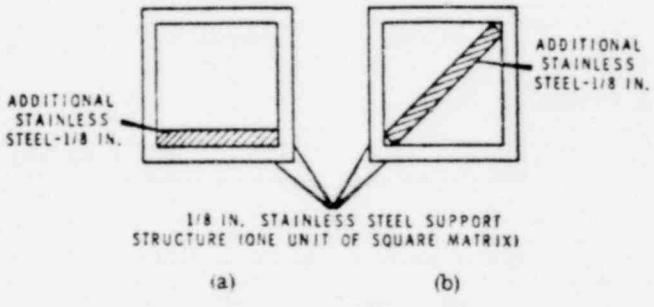

Fig. 2. Support structure matrix with additional steel.

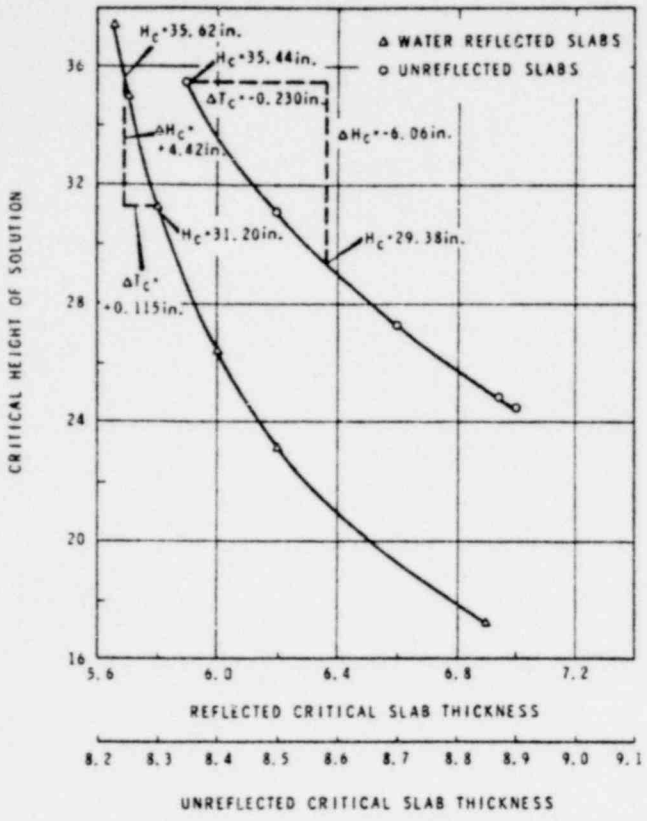

Fig. 3. Stainless steel effects on $Pu(NO_3)_4$ slab experiments.

each of the previously measured slab systems was determined by computing the change in critical thickness for that slab due to the support structure, ΔT^{SS}_{calc}. This value was then converted to the change in the critical thickness which would be measured for the support structure, ΔT^{SS}_{exp}, by assuming that the effects of the support structure could be computed with equal accuracy for all of the critical infinite slab systems. Thus, in a reflected slab $\Delta T^{SS}_{exp} = (0.115/0.104)\Delta T^{SS}_{calc}$, while $\Delta T^{SS}_{exp} = (0.230/0.350)\Delta T^{SS}_{calc}$ for the unreflected systems.

Further experiments were performed to measure the fraction of those neutrons escaping from an unreflected slab that are subsequently returned to the slab from external reflectors such as the hood walls, floor, and the tamper tank framework. This effect was determined by measuring the albedo, β, at the exterior boundary of the support structure using the technique described by Weinstock and Phelps.[18] Since the albedo is a function of position on each face of the slab due to the location of the slab relative to extraneous reflectors, the albedo was measured at positions that should yield the approximate maximum and minimum values on each face of the slab. These measurements indicate that the system-averaged room return is between ~10 and 15%. The average albedo obtained from these measurements is 0.1293 ± 0.0022, or 12.93% room return. Since these measurements do not conclusively yield a system-averaged value for the room return, multiplication factors were computed for the three unreflected systems reported by Lloyd et al.[15,16] as a function of the percent of neutrons returned to the exterior surface of the support structure. The systems considered did not include any corrections for materials extraneous to the solutions. These calculations yield an average room return of 13% as the value necessary to obtain criticality. This result agrees very well with the average measured room return of 12.93%. The critical slab thicknesses were, therefore, computed as a function of the room return for these systems to determine the change in the critical slab thickness associated with a room return of 13%.

The effect of the stainless-steel tank walls on the critical thickness of $Pu(NO_3)_4$ solutions has been computed and measured experimentally. Good agreement has been obtained previously[7] between the computed and measured corrections necessary for both reflected and unreflected spherical systems. It was, therefore, expected that these calculations would yield equally accurate corrections for the slab systems. Measurements[17] show this to be true for the unreflected slabs, but large differences are observed for the reflected slabs. The computed effects of the tank walls for these systems are considered to be valid, however, since not only has agreement with measured values been obtained for other geometries, but the critical thickness of the reflected slabs including the surrounding materials, as well as the individual effect of the support structure, have also been computed accurately. If the measured corrections had been utilized, a maximum change of ~3% in the critical thickness of the clean, infinite slabs would result.

Table I lists the corrections obtained using the above techniques that are necessary to determine the critical thicknesses for clean, infinite slabs of $Pu(NO_3)_4$ solution from the infinite slab data reported by Lloyd et al.[15,16] which include the surrounding materials.

Table II gives the critical thicknesses of the reflected and unreflected infinite slabs of $Pu(NO_3)_4$ solution, including the surrounding system,[15,16] as well as the corrected, clean, critical, infinite slab thicknesses. The computed slab thicknesses are also shown. The maximum difference between a computed and experimental clean, infinite slab thickness is 3%. The average difference between the measured and calculated critical thickness for the seven clean slabs examined is 0.9%.

Using the aforementioned data as a guide, it

TABLE I

$Pu(NO_3)_4$ Solution Slab Tank Corrections

	$Pu(NO_3)_4$ Sol., 2.34*M* HNO_3, 58 g Pu/liter, 4.6% ^{240}Pu (cm)	$Pu(NO_3)_4$ Sol., 5.00*M* HNO_3, 58 g Pu liter, 4.6% ^{240}Pu (cm)	$Pu(NO_3)_4$ Sol., 1.60*M* HNO_3, 202 g Pu/liter, 23.2% ^{240}Pu (cm)	$Pu(NO_3)_4$ Sol., 2.23*M* HNO_3, 284 g Pu/liter, 23.2% ^{240}Pu (cm)
Tank Walls, Reflected	-0.732	-0.757	-0.584	-0.512
Tank Walls, Unreflected	+0.326	+0.336	+0.359	---
Support Structure, Reflected	-0.335	-0.360	-0.476	-0.512
Support Structure, Unreflected	+0.832	+0.866	+0.941	---
Room Return (13%), Unreflected	+1.05	+1.09	+1.24	---

TABLE II

Criticality of Infinite $Pu(NO_3)_4$ Solution Slabs

$Pu(NO_3)_4$ Solution Description	Unreflected Slab Plus System (cm)		Clean-Unreflected Slab (cm)		Water-Reflected Slab Plus System (cm)		Clean Water-Reflected Slab (cm)	
	Exptl.	Calc	Exptl.	Calc	Exptl.	Calc	Exptl.	Calc
2.34*M* HNO_3, 58 g Pu/liter, 4.6% ^{240}Pu	14.7	14.27	16.91	16.91	10.2	10.26	9.13	9.22
5.00*M* HNO_3, 58 g Pu/liter, 4.6% ^{240}Pu	15.7	15.44	17.99	18.19	10.9	11.16	9.78	10.08
1.60*M* HNO_3, 202 g Pu/liter, 23.2% ^{240}Pu	18.65	17.91	21.19	20.94	12.76	12.65	11.70	11.64
2.23*M* HNO_3, 284 g Pu/liter, 23.2% ^{240}Pu	---	---	---	22.05	13.45	13.39	12.43	12.41

appears that the HFN multigroup diffusion theory code, with multigroup constants obtained from the GAMTEC-II code, will compute the critical parameters quite accurately for all clean, one-dimensional plutonium fueled systems containing up to the least 300 g Pu/liter with as much as 25 wt% ^{240}Pu.

Undermoderated Systems

Until recently, with the exception of the plutonium metal experiments,[19] no homogeneous experimental criticality data that could provide a firm basis for establishing criticality safety guidelines have been available for undermoderated systems. The data presently available are extremely limited, but there are a few that can be used as "bench marks" for establishing the accuracy of calculational models in the undermoderated range.

One set of recent experimental data was accumulated using PuO_2-polystyrene plastic compacts at an H/Pu atomic ratio of 15, and having ^{240}Pu isotopic concentrations of 2.2 and 8.08%.[5] These experimental data were acquired for bare and Plexiglas reflected parallelepipeds and were extrapolated by means of buckling conversion to easily defined one-dimensional systems. Critical parameters were computed for these one-dimensional systems using the DTF-IV multigroup transport theory code and were found to agree fairly well with the experimentally determined critical parameters. This calculational method appears to slightly underestimate the critical parameters of the 2.2 wt% ^{240}Pu fuel and, to a slightly greater extent, to overestimate the critical parameters of the 8.08 wt% ^{240}Pu fuel.

The 2.2 wt% ^{240}Pu data have been previously used as a basis for deriving critical parameters of ^{239}Pu(metal)-water mixtures for bare and water reflected systems.[6] Using the same technique, critical parameters for these systems were obtained from the data accumulated on the 8.08 wt% ^{240}Pu fuel. Table III presents a comparison of

TABLE III

Critical Parameters of ^{239}Pu(metal)-Water Mixtures

	Reflector	Predicted Using the 2.2 wt% ^{240}Pu PuO_2-Polystyrene Data (cm)	Predicted Using the 8.08 wt% ^{240}Pu PuO_2-Polystyrene Data (cm)
Infinite Slab Thickness	Bare	11.66 ± 0.30	11.35 ± 0.25
Radius of Sphere	Bare	13.81 ± 0.16	13.41 ± 0.13
Radius of Cylinder	Bare	10.04 ± 0.15[a]	9.77 ± 0.13
Infinite Slab Thickness	Water	4.33 ± 0.07	4.13 ± 0.05
Radius of Sphere	Water	10.40 ± 0.17	10.17 ± 0.14
Radius of Cylinder	Water	6.54 ± 0.14	6.37 ± 0.13

[a]Richey[5] reported a value of 10.52, which recalculation has shown to be incorrect.

these predicted critical parameters. The systematic error introduced by the presence of the additional ^{240}Pu is readily apparent, even though the maximum difference is only 5%. These differences could be caused by 1) the assumption that the experimental data were obtained for homogeneous systems when in reality the system consisted of small PuO_2 particles admixed with polystyrene, or 2) from the incorrect treatment of the resonance absorption by ^{240}Pu in a homogeneous mixture of PuO_2 and polystyrene.

An attempt has been made to determine if the effect of the particulate nature of the PuO_2 is sufficient to account for the disagreement noted between the measured and computed critical parameters reported by Richey[5] for PuO_2-polystyrene mixtures. The effect of having PuO_2 particles embedded in polystyrene, as opposed to having a truly homogeneous PuO_2-polystyrene mixture, are twofold:

1) The depression of the thermal flux in the fuel particle decreases the thermal utilization factor of the material. This effect tends to make the measured critical size of a system containing PuO_2 particles greater than the critical size of an equivalent homogeneous fuel-moderator mixture.

2) The self-shielding of the ^{240}Pu resonance decreases the resonance integral of the mixture, and this effect tends to make the measured critical size of a system containing PuO_2 particles smaller than the size of an equivalent homogeneous fuel-moderator mixture.

The magnitude of these effects increases with the size of the particles. Unfortunately, the particle size distribution was not measured for either of the two oxides used for these experiments; however, an upper limit of 0.15 mm was placed on the diameter of those particles.

To examine the possible magnitude of the effect of the PuO_2 particles, each particle was assumed to be a sphere of the maximum radius, 0.0075 cm. The density of the particles was taken to be the theoretical density of PuO_2, 11.46 g PuO_2/cm^3. These particles were assumed to be embedded in a spherical shell of polystyrene whose density is 1.065 g/cm^3 and whose radius was such that the system-averaged H/Pu atomic ratio was preserved. Since these assumptions do not predict the system-averaged fuel and moderator concentrations, a void region was assumed to be present adjacent to the moderator shell. The assumed dimensions used in the computational model for each ^{240}Pu content fuel are given in Table IV.

TABLE IV

Assumed Dimensions Used in Computational Model

Fuel (wt% ^{240}Pu)	Radius of PuO_2 Particle (cm)	Outer Radius of Polystyrene Shell (cm)	Outer Radius of Void Region (cm)
8.08	7.5×10^{-3}	1.5437×10^{-2}	1.5970×10^{-2}
2.2	7.5×10^{-3}	1.5452×10^{-2}	1.5613×10^{-2}

There is (to our knowledge) no currently available computer code that can accurately evaluate the energy spectrum and, thus, the broad-group material cross sections in a finite system containing multiple, small, but finite heterogenieties. Therefore, the shift in the energy spectrum within the particulate system from that in the corresponding homogeneous mixture could not be evaluated. As a result, it was necessary to assume that the small PuO_2 particles did not cause a spectral shift, and the multigroup cross sections computed for the homogeneous mixtures by the GAMTEC-II code were, therefore, used to compute the spatial effects.

The thermal flux depression within the PuO_2 particle for each ^{240}Pu content fuel was evaluated

using the appropriate thermal-group cross sections. The thermal disadvantage factor, ϕ_M / ϕ_F, was computed for each of the fuel-moderator-void cells. From these values new cell-averaged thermal-group constants were derived for each PuO_2-polystyrene mixture. The adjusted thermal-group constants were lower than the homogeneous values obtained from the GAMTEC-II code by ~3.0% for the 2.2 wt% ^{240}Pu fuel, and by ~3.5% for the 8.08 wt% ^{240}Pu fuel.

To correct for the resonance self-shielding in the particles, Wigner's rational approximation and the narrow resonance infinite mass approximation were assumed valid. The resulting expression for the resonance integral of a particle containing an admixed moderator is

$$I_{(\mathrm{NRIM})} = \int \frac{(\sigma_e + \sigma_m)\,\sigma_{a_F}}{\sigma_{a_F} + \sigma_e + \sigma_m} \frac{dE}{E} ,$$

where the notation is that used by Lamarsh.[20] The cell-averaged cross sections computed for the 2.2 and 8.08 wt% ^{240}Pu fuels based on the above resonance integral decreased from the homogeneous values by about 3 and 5%, respectively. This correction was applied only to the interval 0.683 eV $\leq E \leq$ 1.86 eV, since at higher energies the mean-free-path increased rapidly and, hence, the self-shielding became negligible. [It is important to note that these cross section changes overestimate the true changes by ~1% because the resonance integral calculation does not account for the interaction (Dancoff correction) between the fuel-bearing particles. Since the particle size distribution is not known, however, and since the primary purpose of these calculations was to define the maximum error limits associated with the determination of homogeneous critical parameters from the heterogeneous experimental data, the largest possible effect was the one considered.]

The computed effect of the maximum particle size is small for the 2.2 wt% ^{240}Pu systems, and the corrected critical dimensions of homogeneous PuO_2-polystyrene systems are slightly smaller than the values measured. This is due to the fact that the primary effect was the reduction of the thermal utilization factor. The reflected slab did not follow this trend due to the spectral shift throughout the thin slab caused by the Plexiglas reflector. In this system, the resonance self-shielding was predominant. The effect of the resonance self-shielding was also predominant for all of the systems containing 8.08 wt% ^{240}Pu. As a result, the corrected critical parameters are larger than the values for the measured assemblies. Table V gives an estimate of the corrected homogeneous results obtained by assuming that the average effect of the particle size is the average of the maximum and minimum possible effects (i.e., particle radii of 0.0075 and 0.0 cm). The error limits specified with the estimates of the homogeneous critical parameters incorporate both the maximum and minimum possible effects.

The computed homogeneous critical parameters given in Table V agree with the corrected measured homogeneous critical parameters within

TABLE V

Critical Parameters of PuO_2-Polystyrene Systems at H/Pu = 15

	PuO_2-Polystyrene 2.2% ^{240}Pu			PuO_2-Polystyrene 8.08% ^{240}Pu		
	Experimental[a] (cm)	Experimental[b] (cm)	S_4 Calc[c] (cm)	Experimental[a] (cm)	Experimental[b] (cm)	S_4 Calc[c] (cm)
Bare Arrays						
Infinite Slab Thickness	16.09 ± 0.41	15.96 ± 0.54	15.94	18.45 ± 0.41	18.74 ± 0.67	18.77
Radius of Sphere	18.58 ± 0.22	18.44 ± 0.36	18.25	21.17 ± 0.21	21.44 ± 0.47	21.33
Radius of Infinite Cyl.	13.64 ± 0.21	13.53 ± 0.32	13.34	15.59 ± 0.21	15.79 ± 0.41	15.63
Plexiglas Reflected Arrays						
Infinite Slab Thickness	5.99 ± 0.10	6.03 ± 0.14	6.20	7.35 ± 0.09	7.58 ± 0.29	8.04
Radius of Sphere	13.54 ± 0.22	13.52 ± 0.24	13.47	15.64 ± 0.21	15.89 ± 0.45	15.90
Radius of Infinite Cyl.	8.59 ± 0.19	8.59 ± 0.19	8.53	10.04 ± 0.21	10.22 ± 0.39	10.24

[a]Values derived from experimental data.[5]
[b]Values derived from experimental data and corrected to homogeneous systems.
[c]Values calculated for homogeneous systems.

the true uncertainty of the presently available experimental data. Using these corrected critical parameters to predict the critical parameters of ^{239}Pu(metal)-water mixtures (as was done using the experimental data to obtain the values given in Table III), the agreement between values obtained from the 2.2 and 8.08 wt% ^{240}Pu data is significantly improved. Furthermore, the values obtained using these data agree with the values previously reported by Richey,[6] obtained using the 2.2 wt% ^{240}Pu data, to within 0.06 cm.

Critical experiments have also been performed with PuO_2-polystyrene plastic compacts at an H/Pu atomic ratio of 5 and an isotopic ^{240}Pu concentration of 11.46%.[21] Critical dimensions for clean, one-dimensional systems derived from these data[21] are shown in Table VI along with the corresponding multiplication factors computed by the DTF-IV multigroup transport theory code.

TABLE VI

Critical Parameters of PuO_2-Polystyrene Systems at H/Pu = 5

	Reflector	Critical Dimension (cm)	S_4 Computed k_{eff}
Infinite Slab Thickness	Bare	16.83 ± 0.31	1.0128
Radius of Sphere	Bare	19.68 ± 0.23	1.0198
Radius of Infinite Cylinder	Bare	14.40 ± 0.22	1.0235
Infinite Slab Thickness	Plexiglas	5.98 ± 0.10	0.9635
Radius of Sphere	Plexiglas	14.15 ± 0.37	0.9997
Radius of Infinite Cylinder	Plexiglas	8.93 ± 0.36	0.9998

The particle size distribution was measured for the PuO_2 used for these experiments. The average particle size was found to be 0.0089 mm, with 99% of the particles having a diameter of < 0.02 mm (see Fig. 4). This is much smaller than the maximum particle diameter of 0.15 mm found in the H/Pu = 15 systems. Furthermore, the neutron energy spectrum in the H/Pu = 5 assemblies is much harder than the one in the H/Pu = 15 assemblies. These circumstances reduce the probability of neutrons interacting with the fuel particles. As a result, no corrections were made for the effects of the PuO_2 particles in the H/Pu = 5 systems.

For the reflected infinite slab, the particle size effects may account in part for the observed 3.6% discrepancy in the computed multiplication factor, since for this system some neutrons will be moderated in the reflector and returned to the fuel-bearing region at resonance energies. Since these effects would be small, values of k_{eff} were computed as a function of the order of the S_N approximation to determine if the space-angle mesh assumed in the calculations was adequate to define the thin, undermoderated, reflected infinite slab. These calculations were also made for the reflected infinite slab data reported at an H/Pu ratio of 15, since this same error, to a lesser degree, was also observed for those systems (Table V). The results of these calculations are given in Table VII. As can be seen, the S_4 approximation is sufficiently accurate for most purposes. The primary source of error, therefore, is probably the fact that the cross sections used for the PuO_2-polystyrene region were averaged over a spectrum characteristic of the PuO_2-polystyrene mixture. The true spectrum, however, is significantly more thermal due to the moderation of neutrons in the Plexiglas reflector. This would also account for the fact that the magnitude of the error introduced appears to be a function of the ^{240}Pu content of the fuel.

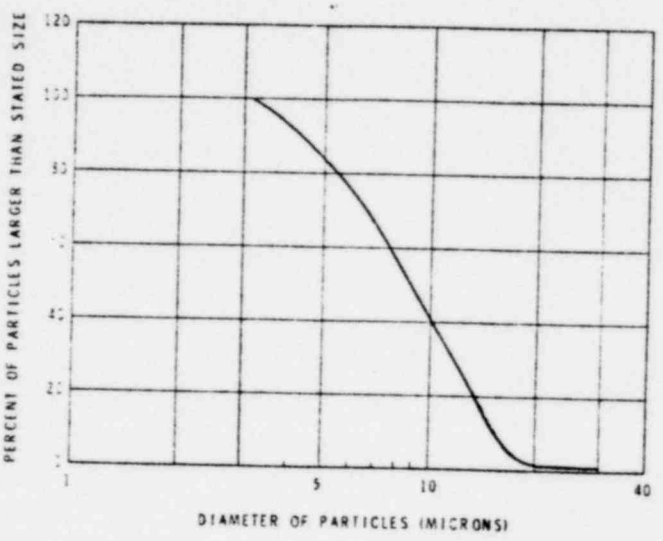

Fig. 4. PuO_2 particle size analysis.

TABLE VII

Computed Multiplication Factors for Plexiglas Reflected Infinite Slabs as a Function of the Order of the S_N Approximation

Fuel	Reported Critical Thickness (cm)	S_4 Computed k_{eff}	S_{16} Computed k_{eff}
PuO_2-Polystyrene H/Pu = 15, 2.2 wt% ^{240}Pu	5.99[a]	0.9895	0.9873
PuO_2-Polystyrene H/Pu = 15, 8.08 wt% ^{240}Pu	7.38[a]	0.9753	0.9732
PuO_2-Polystyrene H/Pu = 5, 11.46 wt% ^{240}Pu	5.98[b]	0.9635	0.9632

[a]Thickness reported by Richey et al.[6]
[b]Thickness reported by Bierman et al.[21]

The error in the computed multiplication factors for unreflected systems represents a discrepancy of ~2% between the measured critical dimensions and the values that would be computed. For example, in the unreflected sphere the difference between the lower limit of the experimentally determined radius and the computed value is only ~0.15 cm. The reflected sphere and infinite cylinder calculations are in excellent agreement.

The critical mass of a water reflected ^{239}Pu sphere has been recently reevaluated.[22] The measured critical mass of 5.43 kg agrees quite well with the value computed using the DTF-IV code, 5.24 kg.

CONCLUSIONS

For well moderated plutonium-fueled systems there are sufficient experimental data to establish the accuracy of the HFN multigroup diffusion theory code when utilizing multigroup constants obtained from the GAMTEC-II code. These calculations are quite accurate for all geometries and for all systems whose isotopic ^{240}Pu content is $\leq 25\%$.

For undermoderated systems there are still very few experimental data on clean, homogeneous systems. The available data, however, do indicate that the critical parameters of reflected homogeneous systems with isotopic ^{240}Pu contents of $\leq 12\%$ can be computed reasonably well using the DTF transport theory code for all geometric configurations except reflected slabs. The reflected slabs cannot be computed accurately using this technique due to the spectral shift which is not accounted for when averaging the energy-dependent cross sections by means of the GAMTEC-II code. For unreflected systems the critical dimensions computed for all geometric configurations deviate from the measured values by no more than the established experimental uncertainties increased by 1% of the critical dimension. It is important to note, however, that larger differences do exist between the computed critical parameters and those that were measured directly. These differences occur in the resonance energy region, notably at H/Pu = 15. The cause of these differences can be defined precisely only through the accumulation and analysis of additional experimental data acquired under very carefully controlled conditions.

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