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Calculations of Criticality Properties of Plutonium Nitrate Systems

W. A. Reardon and J. D. White

Introduction

A series of calculations performed initially as a guide to criticality experiments and to provide the data to check the experiments against the multigroup computational techniques[1] are herein presented. The critical masses, radii, and volumes are computed for aqueous homogeneous plutonium nitrate solutions of 2, 4, and 6 molar nitric acid, and in both bare and reflected spherical geometry. The effect of Pu^{240} is considered by computing the above cases for 0, 2.5, 5, and 7.5% (by-weight) Pu^{240} dilution.

The utility of such calculations is not limited to the checking of experiments, however, since nuclear safety specifications are frequently based upon such calculations. While nuclear safety calculations are commonly based upon idealized solutions of plutonium and water, it is realistic and of considerable interest to compare such calculations with the values obtained with actual nitrate solutions. Heretofore, it has been difficult to perform such calculations accurately, because the chemical compositions were uncertain. This uncertainty coupled with the sensitivity of the calculated parameters to the composition leads to relatively large uncertainties in the computed criticality parameters. The precise determination of the composition of a series of plutonium nitrate solutions has solved this composition problem adequately for plutonium-concentrations under 350 g/l.

Chemical Composition Data

The method of forming a hypothetical plutonium-nitrate solution depends upon the combining of nitric acid of a given molarity with an appropriate amount of the crystal, $Pu(NO_3)_4 \cdot H_2O$(density = 2.56). To form a solution accurately, a plutonium concentration and an acid molarity

are assumed, and the appropriate amounts of the crystal and acid solution are added volumetrically. The resulting atom densities (and volume fractions) of the components are then correct to within a few percent. Higher precision was attained in the calculations presented here by adding enough nitric acid to the solution to force agreement with the measured specific gravity. So far no model for the crystal or the solutions has been completely successful in predicting the composition.

The plutonium solution data are summarized in Table I. *

Basic Computations

Using the chemical data from above, the critical radii were computed, using the 9-Zoom Code[2] and an 18-group set of cross sections.[1] The thermal group cross sections were computed with the TEMPEST[3] Code and substituted for the values on the 9-ZOOM library tape. This procedure provides greater accuracy and flexibility than the 9-ZOOM library alone.

The critical radii for the various amounts of Pu^{240} were computed directly, with a correction to the Pu^{240} resonance integral[4] (assumed to be completely contained in the energy region from 0.625 to 6.0 e. v.). The cross section on the tape is the infinitely dilute value, thus a capture cross section appropriate to the amount of Pu^{240} and the total scattering cross section must be supplied as input. The input data for the computations are summarized in Table II. The range of the calculations is from the region of minimum critical mass to the region of maximum Pu^{240} effect, the expected range of the experiments of the Critical Mass Group of the Hanford Laboratories.[5]

The results of the computations are summarized in Tables III and IV. The critical radii were used to compute the critical volumes, and the volumes together with the plutonium concentrations yield the critical masses.

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* These data were obtained for the Critical Mass Group by W. L. Delvin of the Chemical Development Group of the Hanford Laboratories. They are furnished here, prior to publication with his generous permission.

TABLE I

SPECIFIC GRAVITY VERSUS CONCENTRATION OF Pu, NO_3^-, H^+, and H_2O

(Data from W. L. Delvin)

Sample	Specific Gravity	Pu, g/l	NO_3^-, g/l	H^+, g/l	H_2O, g/l
Stock	1.691	351.8	669.31	5.73	665
A	1.229	31.3	390.5	5.91	797
B	1.271	62.5	416.9	5.66	786
C	1.331	104.0	454.7	5.66	775
D	1.411	154.2	492.5	5.65	760
E	1.468	199.9	531.1	5.45	740
F	1.549	252.6	576.7	5.60	750
G	1.166	30.9	272.3	4	852
H	1.216	61.7	299.7	4	857
I	1.281	102.5	341.4	4	836
J	1.362	156.5	390.5	4	806
K	1.433	206.6	434.4	4	811
L	1.331	31.2	656.7	10	653
M	1.380	62.6	679.2	10	634
N	1.433	102.2	706.2	10	642
P	1.505	155.8	750.2	10	583
Q	1.573	204.1	812.0	10	594
R	1.265	30.9	501.9	7.5	714
S	1.313	61.1	525.8	7.5	727
T	1.370	102.2	565.5	7.5	711
U	1.445	153.0	600.6	7.5	703
V	1.516	205.0	658.1	7.5	671
W	1.583	255.2	695.0	7.5	632
X	1.138	49.7	174.3	2	950
Y	1.184	79.6	196.3	2	954
Z	1.258	125.0	247.3	2	927
AA	1.074	29.2	91.0	1	989
BB	1.120	59.7	118.3	1	979

TABLE II

COMPOSITION AND THERMAL GROUP PARAMETERS

H:Pu	Plutonium g/l	Specific Gravity	H_2O g/l	NO_3^- g/l	1/v σ_a	239 σ_c	$239^{(1)}$ σ_f	H. σ_{tr}
2M HNO_3								
836	30	1.112	926.5	153.0	0.746	334.7	705.3	29.5
499	50	1.146	921.4	173.0	0.715	347.4	706.6	28.3
354	70	1.182	916.7	193.0	0.689	355.3	705.7	27.2
274	90	1.216	911.4	212.8	0.665	361.8	703.2	26.3
161	150	1.316	891.8	271.9	0.607	369.8	696.2	23.9
92.6	250	1.477	855.7	369.9	0.544	368.4	664.6	21.3
53.7	400	1.712	794.7	516.1	0.475	350.2	617.7	18.5
31.5	600	2.009	698.8	708 8	0.420	322.6	564.3	16.3
25.1	700	2.160	650.9	807.8	0.401	309.3	540.8	15.5
4M HNO_3								
792	30	1.167	859.4	273.2	0.704	360.7	715.3	27.8
473	50	1.203	856.5	292.8	0.678	368.2	713.8	26.8
335	70	1.235	850.1	311.4	0.654	373.4	710.6	25.8
259	90	1.266	844.1	330.0	0.633	376.7	706.3	25.0
152	150	1.386	827.2	386.2	0.582	378.3	689.7	22.9
87.2	250	1.520	789.6	477.0	0.525	372.1	660.9	20.6
50.2	400	1.743	727.2	612.1	0.462	349.4	611.1	18.0
29.2	600	2.028	625.7	789.3	0.411	319.5	556.5	15.9
23.2	700	2.172	589.1	880.4	0.392	305.4	532.6	15.1
6M HNO_3								
752	30	1.227	796.6	394.3	0.666	384.6	724.7	26.3
448	50	1.250	785.4	409.3	0.643	387.7	720.3	25.4
315	70	1.283	780.5	427.2	0.622	389.1	714.9	24.5
244	90	1.317	776.3	445.4	0.604	390.2	708.8	23.8
143	150	1.410	757.5	496.9	0.559	386.5	688.5	22.0
81.7	250	1.559	722.3	581.2	0.507	374.4	656.6	19.9
46.8	400	1.772	622.5	704.4	0.449	348.1	604.3	17.5
27.0	600	2.044	575.5	865.0	0.402	316.0	548.7	15.5
21.3	700	2.182	531.1	947.6	0.385	301.4	524.5	14.8

TABLE III

CALCULATED CRITICAL PARAMETERS FOR PLUTONIUM-NITRATE SOLUTIONS HOMOGENEOUS BARE SPHERES

		0% Pu^{240}			2.5% Pu^{240}			5% Pu^{240}			7.5% Pu^{240}		
H/Pu	Acid	R_c, cm	V_c, l	M_c, kg	R_c, cm	V_c, l	M_c, kg	R_c, cm	V_c, l	M_c, kg	R_c, cm	V_c, l	M_c, kg
836	2.0	21.10	39.35	1.18	21.72	42.92	1.255	22.26	46.20	1.317	22.82	49.78	1.381
499	2.0	18.87	28.14	1.41	19.40	30.58	1.491	19.87	32.88	1.562	20.33	35.21	1.629
354	2.0	17.94	24.19	1.69	18.57	26.86	1.833	19.03	28.87	1.926	19.43	30.73	1.990
274	2.0	17.52	22.53	2.03	18.15	25.04	2.197	18.63	27.08	2.316	19.06	29.00	2.414
161	2.0	16.92	20.29	3.04	17.65	23.03	3.367	18.19	25.20	3.591	18.64	27.12	3.763
92.6	2.0	16.53	18.92	4.73	17.44	22.22	5.416	18.04	24.60	5.843	18.56	26.60	6.151
53.7	2.0	16.28	18.07	7.23	17.30	21.70	8.463	17.93	24.15	9.177	18.39	26.05	9.639
31.5	2.0	16.35	18.31	10.98	17.40	22.05	12.899	18.40	24.59	14.017	18.55	26.72	14.830
25.1	2.0	16.40	18.48	12.93	17.45	22.25	15.186	18.06	24.67	16.408	18.68	27.30	17.077
792	4.0	21.86	43.76	1.31	22.45	47.39	1.386	23.04	51.23	1.460	23.60	55.05	1.528
473	4.0	19.51	31.11	1.55	20.09	33.96	1.655	20.59	36.55	1.737	21.06	39.13	1.810
335	4.0	18.64	27.13	1.96	19.23	29.80	2.034	19.73	32.17	2.139	20.18	34.44	2.239
259	4.0	18.14	25.00	2.25	18.80	27.83	2.442	19.32	30.20	2.668	19.75	32.26	2.685
152	4.0	17.45	22.26	3.34	18.23	25.38	3.711	18.82	27.92	3.979	19.22	29.72	4.124
87.2	4.0	17.07	20.83	5.21	18.02	24.51	5.974	18.68	27.28	6.479	19.07	29.03	6.714
50.2	4.0	16.87	20.11	8.04	17.96	24.27	9.463	18.60	26.95	10.243	19.20	29.65	10.970
29.2	4.0	16.93	20.33	12.20	18.07	24.71	14.458	18.71	27.43	15.638	19.23	29.80	16.539
23.2	4.0	17.04	20.72	14.51	18.14	25.00	17.064	18.78	27.74	18.450	19.28	30.04	19.451
752	6.0	22.54	47.97	1.44	23.18	52.17	1.526	23.79	56.40	1.607	24.34	60.60	1.612
448	6.0	20.30	35.04	1.75	20.93	38.41	1.872	21.48	41.52	1.972	22.06	44.60	2.069
315	6.0	19.35	30.35	2.12	20.02	33.61	2.294	20.58	36.50	2.427	21.09	39.30	2.545
244	6.0	18.80	27.83	2.50	19.55	31.30	2.746	20.08	33.95	2.903	20.55	36.35	3.027
143	6.0	18.09	24.80	3.72	18.96	28.55	4.176	19.56	31.35	4.468	20.06	33.80	4.690
81.7	6.0	17.67	23.11	5.78	18.70	27.39	6.677	19.35	30.35	7.208	19.88	32.90	7.608
46.8	6.0	17.48	22.37	8.95	18.61	27.00	10.529	19.29	30.07	11.427	19.85	32.75	12.093
27.0	6.0	17.58	22.76	13.65	18.75	27.61	16.153	19.34	30.30	17.271	19.97	33.60	18.748
21.3	6.0	17.75	23.42	16.40	18.85	28.06	19.492	19.49	31.01	20.623	20.05	33.76	21.828

TABLE IV

CALCULATED CRITICAL PARAMETERS FOR PLUTONIUM-NITRATE SOLUTIONS HOMOGENEOUS WATER REFLECTED SPHERES*

		0% Pu^{240}			2.5% Pu^{240}			5% Pu^{240}			7.5% Pu^{240}		
H:Pu	Acid	R_c, cm	V_c, l	M_c, kg	R_c, cm	V_c, l	M_c, kg	R_c, cm	V_c, l	M_c, kg	R_c, cm	V_c, l	M_c, kg
836	2.0	17.06	20.76	0.623	17.65	23.05	0.675	18.13	24.96	0.711	18.66	27.20	0.755
499	2.0	14.80	13.58	0.679	15.29	14.97	0.730	15.71	16.24	0.771	16.11	17.52	0.810
354	2.0	13.88	11.20	0.784	14.41	12.53	0.855	14.73	13.39	0.809	15.06	14.30	0.926
274	2.0	13.44	10.17	0.915	13.99	11.47	1.006	14.36	12.40	1.060	14.68	13.25	1.104
161	2.0	12.80	8.78	1.32	13.40	10.08	1.474	13.79	10.98	1.566	14.25	12.12	1.682
92.6	2.0	12.42	8.02	2.01	13.31	9.88	2.470	13.72	10.82	2.569	14.09	11.72	2.709
53.7	2.0	12.25	7.69	3.08	13.07	9.35	3.647	13.63	10.61	4.030	14.07	11.67	4.316
31.5	2.0	12.27	7.74	4.64	13.17	9.57	5.597	13.74	10.86	6.193	14.15	11.87	6.586
25.1	2.0	12.34	7.87	5.51	13.23	9.70	6.620	13.80	11.01	7.321	14.21	12.02	7.782
792	4.0	17.62	22.93	0.688	18.20	25.25	0.758	18.75	27.59	0.828	19.25	29.90	0.830
473	4.0	15.35	15.15	0.757	15.85	16.68	0.813	16.32	18.21	0.864	16.74	19.65	0.909
335	4.0	14.42	12.56	0.879	14.98	14.08	0.967	15.42	15.36	1.021	15.82	16.58	1.074
259	4.0	13.93	11.32	1.02	14.52	12.82	1.125	14.96	14.02	1.199	15.36	15.18	1.264
152	4.0	13.21	9.66	1.45	13.89	11.22	1.642	14.34	12.35	1.760	14.73	13.39	1.857
87.2	4.0	12.85	8.89	2.22	13.64	10.63	2.590	14.18	11.94	2.837	14.55	12.90	2.983
50.2	4.0	12.67	8.52	3.41	13.55	10.42	4.064	14.08	11.69	4.443	14.55	12.90	4.773
29.2	4.0	12.70	8.58	5.15	13.64	10.63	6.210	14.18	11.94	6.808	14.67	13.22	7.339
23.2	4.0	12.79	8.76	6.13	13.70	10.77	7.351	14.30	12.25	8.145	14.76	13.47	8.723
752	6.0	18.25	25.46	0.764	18.83	27.97	0.818	19.42	30.68	0.874	19.95	33.26	0.923
448	6.0	15.97	17.06	0.853	16.50	18.82	0.917	16.98	20.50	0.974	17.42	22.14	1.024
315	6.0	14.98	14.08	0.986	15.57	15.81	1.079	16.02	17.22	1.145	16.46	18.68	1.210
244	6.0	14.45	12.64	1.14	15.07	14.34	1.258	15.56	15.78	1.349	15.97	17.06	1.421
143	6.0	13.71	10.79	1.62	14.43	12.59	1.841	14.98	14.08	2.006	15.36	15.18	2.106
81.7	6.0	13.29	9.83	2.46	14.11	11.77	2.868	14.67	13.22	3.305	15.14	14.54	3.361
468	6.0	13.10	9.42	3.77	14.02	11.54	4.501	14.63	13.12	4.984	15.07	14.33	8.165
21.3	6.0	13.25	9.74	6.82	14.19	11.97	8.168	14.79	13.55	9.012	15.28	14.95	9.677

* Water reflector thickness of one foot is assumed in all cases.

Derived Quantities

From the above computed quantities can be estimated the effects of some materials of interest in nuclear safety specifications.

The Effect of Pu^{240} Concentration

The effect of the increments of Pu^{240} concentration are shown in Figure 1. Two effects show up, not unexpectedly, concerning the incremental concentration of Pu^{240} and the acid.

a. Each added increment of Pu^{240} in the solution has less effect than the previous increment. This effect is expected on a physical basis by considering that in a light water system a certain fraction of the neutrons slowing down in the energy region of the large Pu^{240} resonance will escape capture by scattering past the resonance on the first collision upon entering the region. The probability of the remainder escaping capture will depend upon the ratio of the total scattering and capture cross sections, the capture cross section being very large in the vicinity of the resonance. The number of neutrons being captured, however, is limited to those failing to scatter past the resonance on the first collision. If a given amount of Pu^{240} captures a large fraction of the neutrons in the energy range, then adding more Pu^{240} can only capture some fraction of the small fraction remaining. This representation is, of course, over simplified, but it provides the intuitional basis for the diminution of the Pu^{240} effect. Mathematically, the probability of capture in a single large resonance is proportional to:

$$(1)\quad p_j \sim \frac{1}{\xi \Sigma_s}\left(\frac{N^{240}\sigma_c}{\sqrt{1+\frac{\sigma_r}{\sigma_{sn}}}}\right)$$

$$\text{or for large } N^{240} \quad = \frac{1}{\xi \Sigma_s}\left(\sqrt{N^{240}\sigma_c}\,\sigma_r\sqrt{\sigma_{sn}}\right).$$

where: ξ = the average logrithmic energy decrement.

Σ_s = the total moderator scattering cross section,

$\sigma_r = \sigma_c + \sigma_s$, maximum value of the total cross section of Pu^{240} at the resonance,

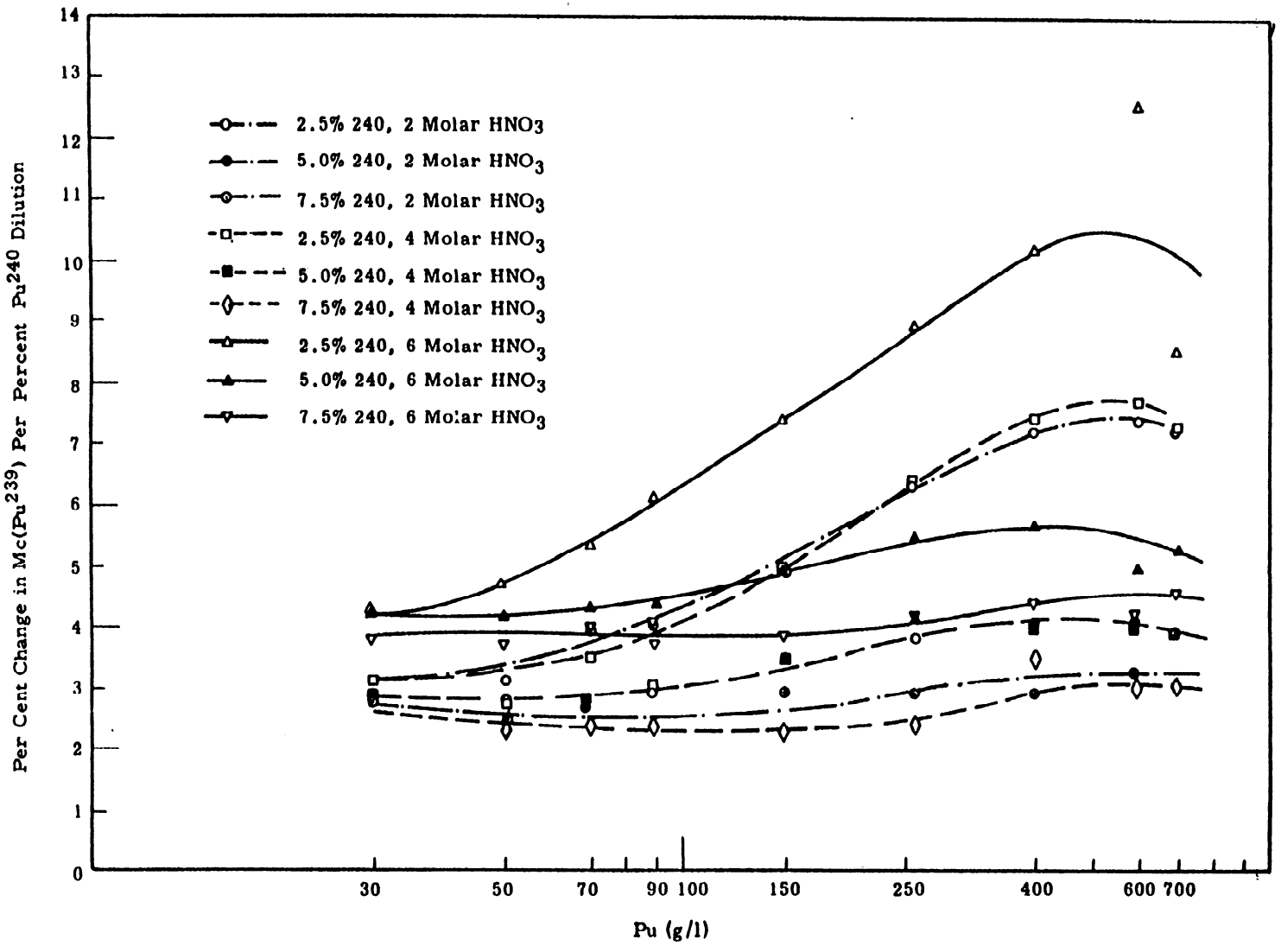

FIGURE 1

Incremental Pu^{240} Effect in Bare Spheres

σ_{sn} = the number of scattering barns per absorber atom

$$= \frac{\sum N^i \sigma_s^i}{N^{240}} \quad \text{and}$$

N^i = number density of atoms of the i'th isotope

σ_s^i = microscopic scattering cross section of the i'th isotope,

and N^{240} = number density of Pu^{240} atoms.

From the above equation, it is seen that the absorption probability increases only as the square-root of the Pu^{240} concentration.

b. The second effect, that the effect of Pu^{240} as an absorber increases with increasing nitrate concentration, can also be surmised on intuitional grounds, to wit: Neutrons scattering from hydrogen will on the average lose one-half of their energy in each scatter and thus have a fixed, finite probability of "scattering past" the resonance during the slowing down process. If the hydrogen is displaced by an atom for which the energy loss by a neutron, per collision is approximately 5 percent, then the neutron must suffer approximately 10 times as many collisions to lose one-half its energy. The neutron then has approximately 10 times the opportunity to suffer a capture collision. Again this "intuition" is over-simplified, but equation 1 again demonstrates the point. The fraction in front of the equation, when the moderator is a mixture of isotopes, is given by:

$$\frac{1}{\bar{\xi}\,\bar{\Sigma}_s} = \frac{1}{\sum_i \xi_i \Sigma_i^s} = \frac{1}{\sum_i \xi_i N^i \sigma_s^i}$$

(Refer to definitions with equation 1.)

If the moderator were principally hydrogen, then the dominant part of the denominator is $(\xi_h N^h \sigma_s^h)$. In the case of the nitrate mixtures

$$\left.\begin{aligned} \xi_h &> \xi_j \\ \sigma_s^h &> \sigma_s^i \end{aligned}\right\} (i \neq h)$$

Increasing the concentration of nitrogen and oxygen by adding (NO_3) to the system causes the denominator to get smaller and the probability of capture to increase. Also, the denominator in the square root term will decrease, but its effect is only as the square root of the total scattering cross section, thus the effect of Pu^{240} will increase as the hydrogen is displaced by heavier isotopes.

The Effect of Nitrate-ion Concentration

The displacement of H_2O by (NO_3) can be expected to have three separate effects upon the critical mass (or volume or radius):

a. The effect of the probability of capture by Pu^{240} as outlined in part b above.
b. The capture cross section of nitrogenis larger than that of hydrogen and will thus act as a poison when displacing hydrogen.
c. Because the nitrogen has a higher capture cross section than hydrogen it will cause the thermal neutron energy spectrum to be "hardened" and be shifted in the direction of a higher capture-to-fission ratio (α) in the Pu^{239} cross section.

Effects (b) and (c) occur together, but the effect (α) can be partly isolated by varying the Pu^{240} content of the solution.

Figure 2 shows the dependence of the critical volume on the incremental changes in (NO_3^-) concentration, expressed as the change in acid molarity for 0 and 5 percent Pu^{240} content. One observes that in this case the second increment is more effective than the first. This might be expected on the basis that the spectral shift is more effective as a poison than the added absorption in the thermal group. The total effect, however is approximately a factor of three smaller in the case of reflected systems, and thus a change in leakage is probably a larger effect still. This would result from a lowering of the total scattering cross section at the higher acid concentrations.

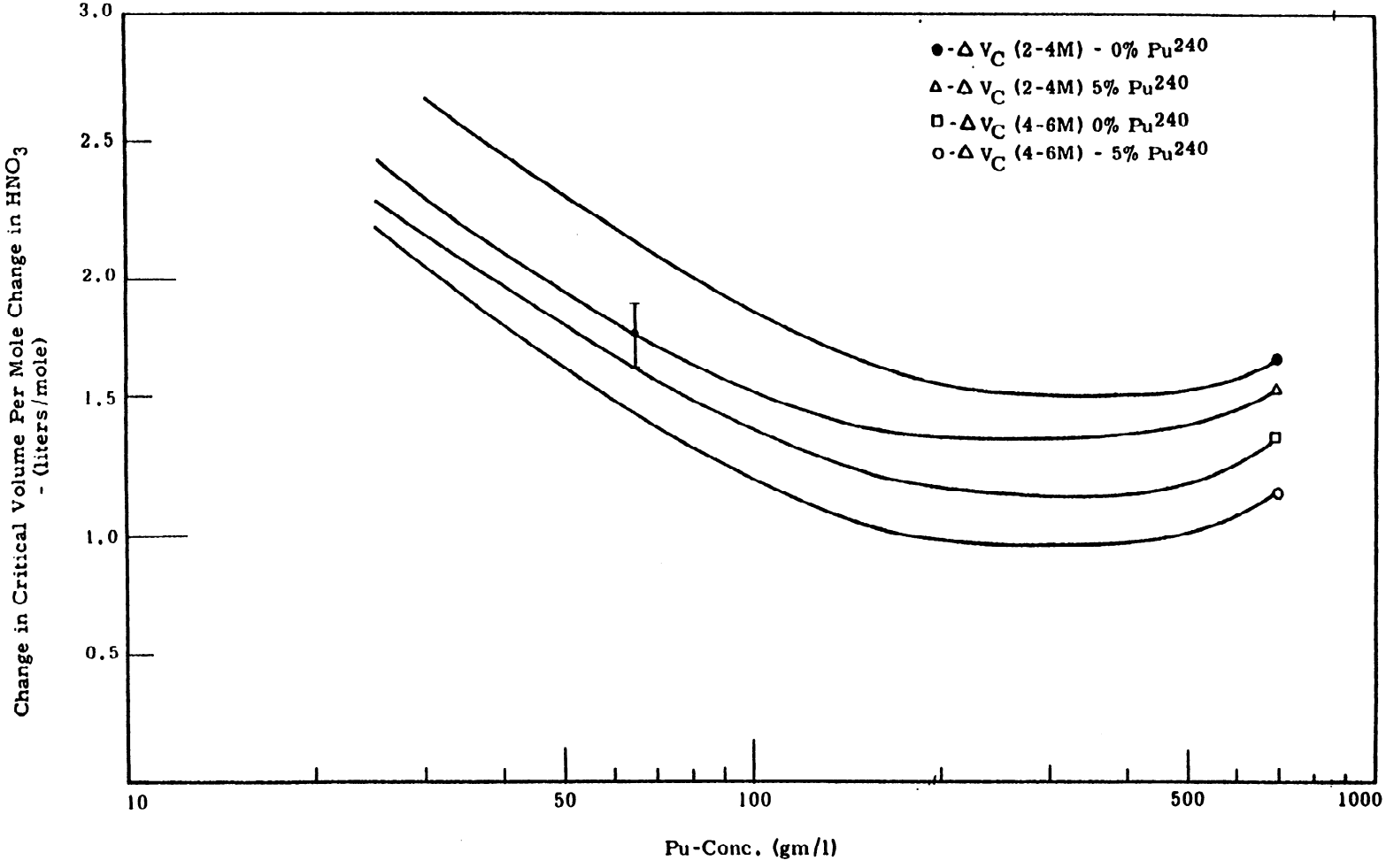

FIGURE 2

Incremental Nitrate-ion Effect in Bare Spheres

Estimates of Precision

The 9-Zoom code converges on the effective multiplication constant as the eigenvalue for a given composition and geometry. The calculations were performed by running a series of problems of different radii and fixed composition. The convergence limits used were adequate to assure accurate values of the "k_{eff}" and the principal functional uncertainty arises from the interpolation of the radii to secure the critical radius. The radii were generally computed to the nearest 0.05 cm and can be uncertain by as much as ± 0.1 cm at the lower plutonium concentrations. This leads to an uncertainty of the order of 0.5 liters in the critical volume, and uncertainties in the critical masses of from 15 grams at 30 g/l to 200 grams at 600 g/l. These uncertainties are supplemented by the uncertainties in the cross sections used in the calculation. It has been impossible to isolate and estimate accurately their individual effects. Refuge is therefore taken in criticality data and a comparison between measured and computed values of particular parameters of various systems. The cross sections with 9-Zoom (though the code is probably blameless) generally underestimate the critical masses of U^{235}-H_2O systems by ~ 2 to 4% over a wide range of H/U ratios.(6) The evidence is not complete for plutonium-H_2O systems, but it appears that the critical masses in the range of 25 to 100 g/l plutonium concentration are uncertainly computed by ± 2% depending upon the composition.(7) Later data obtained at the Hanford Critical Mass Laboratory(8) indicate that the agreement is good from 100 to 300 g/l and high acid concentrations, but are inconclusive at lower concentrations. Corrections for Pu^{240} and HNO_3 concentrations as computed also agree within the experimental uncertainties with data so far obtained.

The computed incremental effects of Pu^{240} and (NO_3) concentrations suffer from a lack of significant figures, because the convergence criterion of the radius was not particularly tight. In the case of the Pu^{240} correction some smoothing of the data was possible, without effecting the uncertainty, by cross plotting the data in Table III. Such cross plotting helps eliminate

internal inconsistencies and forces a careful proof reading; the quality of the results is thereby improved. The scatter in the points on the curves indicates the extent of the uncertainties. The same source of uncertainties applies in the case of the incremental $(NO_3)^-$ concentration effect. In this case the smoothed data were used and the uncertainty arising from the radii uncertainty is shown by the vertical bars on Figure 2. Some revision of the data can be expected as more experimental data become available.

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