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The Measurement of Eta and the Limiting Concentration of ^{239}Pu in Critical Aqueous Solutions

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Experiments were performed with a 122-cm-diam sphere to determine criticality of aqueous solutions of plutonium in a system having low-neutron leakage. The plutonium in the chemical form of $Pu(NO_3)_4$ had a ^{240}Pu content of 2.52 wt%. The critical-sphere concentration obtained in this experiment was analyzed along with data from eight additional critical experiments to evaluate the minimum critical concentration for plutonium. The limiting critical concentration was determined to be 7.62 g Pu/ℓ for $Pu(NO_3)_4$ without excess acid and 7.59 g Pu/ℓ for a ^{239}Pu-water mixture. From these data, the Maxwellian-averaged thermal value of the number of fission neutrons emitted per neutron absorbed by ^{239}Pu, eta, was determined to be 2.056 ± 0.037. The value at 2200 m/s is 2.100 ± 0.041.

I. INTRODUCTION

Experiments have been performed to provide data for determining the thermal value of the number of fission neutrons emitted per neutron absorbed by ^{239}Pu, eta, and the limiting critical concentration of ^{239}Pu in an aqueous solution. The limiting critical concentration is that concentration of the fissile nuclide in solution at which the infinite reproduction factor (k_∞) is unity. At lower concentrations, the reproduction factor would be less than unity for systems of any size or shape. This value will be useful as a benchmark for testing calculation methods and cross-section sets in well-thermalized systems.

In previous work, the limiting critical concentration was determined indirectly from "null reactivity" measurements on a relatively small volume of solution, wherein several corrections and extrapolations were required to determine the concentration.[1] Analysis of the data gave a value of 7.8 to 8.0 g Pu/ℓ. A later evaluation of these data produced a value of 7.2 g/ℓ (Ref. 2).

Criticality experiments have been performed previously on small spheres with high-plutonium concentration.[3] The present experiments were performed using a very large (4-ft-diam) sphere to obtain a low-leakage plutonium system suitable for determining the thermal value of eta.

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[1]R. H. MASTERSON, J. D. WHITE, and T. J. POWELL, "The Limiting Critical Concentrations for ^{239}Pu and ^{235}U in Aqueous Solutions," HW-77089, Hanford Atomic Products Operation (1963).

[2]C. R. RICHEY, *Trans. Am. Nucl. Soc.*, **17**, 279 (1973).

[3]R. C. LLOYD, C. R. RICHEY, E. D. CLAYTON, and D. R. SKEEN, *Nucl. Sci. Eng.*, **25**, 165 (1966).

A discussion of these experiments was presented[4] previously, and a copy of that presentation was distributed. This paper gives a more detailed analysis of the experimental data using better estimates of the plutonium concentration of the experimental solutions.

Also, the data from this experiment were used as the basis for the subcritical limit on concentration[5] being proposed in a revision of the American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors.[6]

II. EXPERIMENTAL METHOD

The 4-ft-diam aluminum sphere is shown in Fig. 1. This sphere was previously used in critical experiments at the Oak Ridge National Laboratory (ORNL) in the measurement of eta for uranium in critical aqueous solutions.[7] The plutonium in the current experiments was selected for its low ^{240}Pu content of 2.52 wt%. The sphere volume was 949.1 ℓ. For low-plutonium concentrations in aqueous solutions, the neutron leakage from a sphere of this size will be no more than a few percent. A series of measurements was made with $Pu(NO_3)_4$ solutions with plutonium concentrations ranging from 11 to <9.5 g Pu/ℓ with nitric acid molarity near unity.

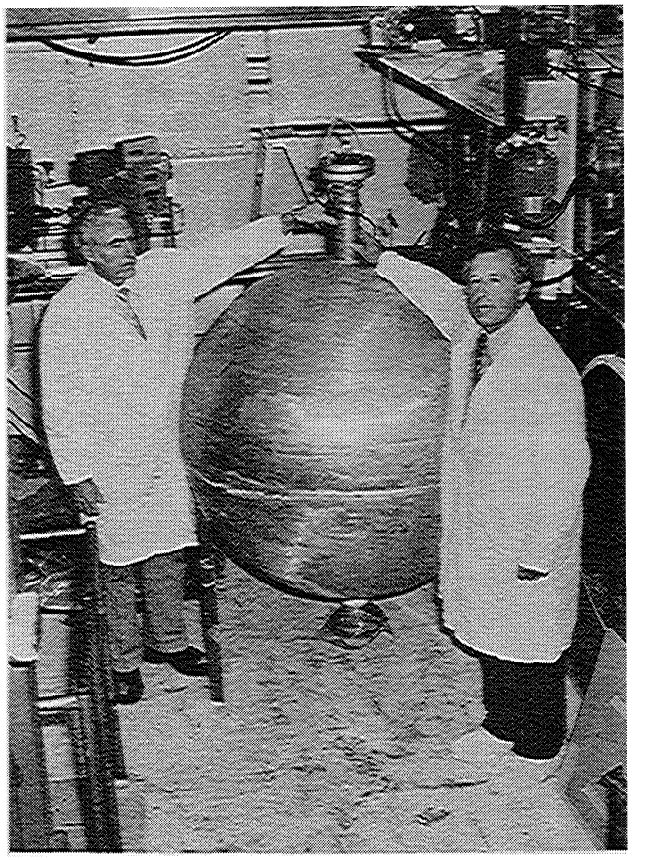

Fig. 1. Large aluminum sphere used in eta experiment.

III. THEORETICAL BASIS

The critical equation for an aqueous homogeneous thermal system can be written as

$$k_{eff} = k_{\infty} P(B) = 1 \quad , \tag{1}$$

in which $P(B)$ is the nonleakage probability and the infinite reproduction factor $k_{\infty} = \bar{\eta} f F$. The factor F accounts for epithermal fission and absorption of neutrons while slowing down; f denotes the common thermal utilization. As the system is increased in size, the neutron leakage becomes small, and $P(B)$ will approach unity. Under these circumstances, $\bar{\eta}$ (apart from small corrections for epithermal fissions) will become equal to $1/f$ (the reciprocal of the thermal utilization). The measurement of eta then depends on

1. knowledge of absorption cross sections for the aqueous solutions in question in order to compute f
2. the nonleakage probability, which must be either computed or measured.

A dilute homogeneous aqueous solution was used so that the critical size would be large [$1 - P(B)$ small] and the neutrons would be well thermalized. Both conditions are required for an accurate measurement of eta. Small neutron leakage means less dependence on the accuracy of either the measured or calculated values of the critical (geometrical) buckling, the age, and thermal diffusion length; thermal neutron absorption cross sections are relatively well known.

The value of eta from Eq. (1) is given by

$$\bar{\eta} = \frac{k_{eff}}{fFP(B)} \quad . \tag{2}$$

For the critical sphere we obtain

$$\bar{\eta} = \frac{1}{fFP(B)} \quad , \tag{3}$$

[4]R. C. LLOYD, R. A. LIBBY, and E. D. CLAYTON, *Trans. Am. Nucl. Soc.*, **28**, 292 (1978).

[5]H. K. CLARK, *Nucl. Sci. Eng.*, **79**, 65 (1981).

[6]"American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," N16.1-1975 (ANS-8.1), American Nuclear Society (1975).

[7]R. GWIN and D. W. MAGNUSON, *Nucl. Sci. Eng.*, **12**, 364 (1962).

TABLE I

Experimental Results

Experiment Number	Extrapolated[a] Critical Volume (ℓ)	Plutonium Concentration[b] (g/ℓ)	HNO_3 Molarity	Specific Gravity
3	656.6 ± 0.5	10.02	1.10	1.055
5	906.5 ± 0.1	9.539 ± 0.033	1.10	1.053
7	991.8 ± 0.8	9.374 ± 0.048	1.11	1.053
Interpolated critical	949.1 ± 5.3	9.457 ± 0.029	1.105 ± 0.01	1.053 ± 0.005

[a]The critical volume was determined by extrapolation of the inverse multiplication curves near criticality (vessel contained >95% of the critical mass).

[b]The plutonium concentrations were chosen from two sets of chemical analyses. Although the two sets of values were in good agreement, one set was discarded because of problems later disclosed with the chemical analysis technique.

where

$$F = \frac{\bar{\Sigma}_{ax}}{\bar{\Sigma}_{at}}$$

in which $\bar{\Sigma}_{ax}$ is the average thermal macroscopic absorption cross section of the fuel, and $\bar{\Sigma}_{at}$ is the average total thermal macroscopic absorption cross section. Furthermore,

$$F = \epsilon p$$

in which ϵ is the fast fission factor and p is the resonance escape probability;

$$P(B) = \frac{1}{(1 + L^2B^2)(1 + \tau B^2)} ,$$

where

L = diffusion length

B^2 = critical buckling

τ = Fermi age.

Values for these parameters are calculated from data in Sec. IV.

IV. EXPERIMENTAL RESULTS

The data from the critical approaches on the large sphere, and the chemical analyses for the associated solutions, are given in Table I. The capacity of the aluminum sphere was taken to be 949.1 ℓ, based on the following ORNL description:

"Hemispherical shells die-formed from type 1100 aluminum plate were welded to form the sphere. Flanged tubes 3 in. [7.62 cm] in diameter were welded at the poles. Sonic measurements of the wall thickness ranged from 0.26 to 0.35 in. [0.66 to 0.89 cm]. The average thickness, based on the weight of aluminum in the shells, was 0.303 in. [0.77 cm]. The capacity, 949.1 liters, was measured by weighing the water contained by the sphere. The volume-averaged diameter was 48.04 in. [122.02 cm]. The outside diameter was measured at 28 places and found to be 48.65 ± 0.09 in. [123.57 ± 0.23 cm]. Maximum and minimum values were 48.87 and 48.50 in. [124.13 and 123.19 cm]. The interior surfaces were coated with a phenolic base baked-on enamel to prevent corrosion."

The critical concentration for the full sphere was determined by a linear interpolation between experiments 5 and 7. The plutonium concentrations determined by chemical analyses at the critical volumes are also shown in Table I.

Table II gives the plutonium isotopic distribution. The elemental impurities are shown in Table III. All experiments were performed at a temperature of 23 ± 0.5°C.

V. ETA EVALUATION

The atom densities for the critical spherical solution are shown in Table IV. The thermal neutron

TABLE II

Plutonium Isotopic Distribution

Isotope	wt%
^{238}Pu	0.004 ± 0.001
^{239}Pu	97.386 ± 0.003
^{240}Pu	2.521 ± 0.001
^{241}Pu	0.075 ± 0.001
^{242}Pu	0.014 ± 0.001

TABLE III

Chemical Impurities

Element	Concentration by Weight Relative to Plutonium (ppm)
Aluminum	10 000
Barium	10
Beryllium	4
Boron	14
Bromine	10
Cadmium	7
Calcium	300
Chlorine	200
Chromium	1 500
Cobalt	10
Copper	50
Dysprosium	7
Europium	10
Fluorine	1 000
Gadolinium	10
Gallium	5 000
Germanium	3
Iron	7 000
Lead	35
Lithium	6
Magnesium	200
Manganese	150
Molybdenum	15
Nickel	1 000
Phosphorus	1 000
Potassium	100
Samarium	20
Silicon	500
Silver	10
Sodium	300
Sulfur	1 000
Tantalum	50
Tin	15
Titanium	20
Tungsten	68
Uranium-235	300
Vanadium	10
Zinc	50
Zirconium	30

TABLE IV

Atom Densities in Critical Solution

Isotope	Atom Density (atom/b·cm)
^{238}Pu	$(1.077 \pm 0.239) \times 10^{-9}$
^{239}Pu	$(2.3202 \pm 0.0071) \times 10^{-5}$
^{240}Pu	$(5.9813 \pm 0.0218) \times 10^{-7}$
^{241}Pu	$(1.7720 \pm 0.0242) \times 10^{-8}$
^{242}Pu	$(3.294 \pm 0.239) \times 10^{-9}$
Hydrogen	$(6.4844 \pm 0.0280) \times 10^{-2}$
Nitrogen	$(7.6078 \pm 0.0603) \times 10^{-4}$
Oxygen	$(3.4372 \pm 0.0141) \times 10^{-2}$

cross sections used in the calculations are from ENDF/B-IV (Ref. 8) and are listed in Table V. The parameters used in the calculation of eta using Eq. (3) are given in Table VI. The fast fission factor and the effect of absorption of neutrons while slowing down were derived from calculations by the EGGNIT code.[12] Values for the age and L^2 were also obtained from EGGNIT.

The value of eta that results from the critical data is 2.056 ± 0.037. This corresponds to a 2200 m/s value of 2.100 ± 0.041, which compares to the presently accepted value of 2.111 ± 0.008 (Ref. 11). A sensitivity analysis was made to determine the major source of uncertainty in the calculated value of eta. The largest single source of error is the uncertainty in the thermal fission cross section for ^{239}Pu. The value used was 754.84 ± 4.5 b from Leonard and Thompson.[11] This uncertainty alone is equivalent to an uncertainty in eta of ±0.029 and of η(2200 m/s) of ±0.033. The uncertainty in $\bar{\eta}$ due to uncertainties in atom densities (including uncertainties in the critical concentration, isotopic distribution, nitrate concentration, and plutonium concentration) is ±0.016 and for η(2200 m/s) is ±0.017. The uncertainty due to variation in extrapolation length for the buckling calculations amounts to ±0.013 in $\bar{\eta}$ and ±0.014 for η(2200 m/s).

VI. MINIMUM CRITICAL CONCENTRATION

The minimum critical concentration for plutonium was determined by first calculating k_{eff} for a

[8]D. I. GARBER and C. BREWSTER, "ENDF/B Cross Sections," BNL-17100 (ENDF-200), 2nd ed., Brookhaven National Laboratory (1975).

[9]J. W. BOLDEMAN, "Review of $\bar{\nu}$ for ^{252}Cf and Thermal Neutron Fission," *Neutron Standards and Applications, Proc. Int. Specialists' Symp. Neutron Standards and Applications*, Gaithersburg, Maryland, March 28-31, 1977, NBS Special Publication 493, p. 182, U.S. National Bureau of Standards (1977).

[10]C. WAGEMANS and A. J. DERUYTTER, *Annals of Nuclear Energy*, Vol. 2, p. 541, Pergamon Press (1975).

[11]B. R. LEONARD, Jr. and J. K. THOMPSON, "Evaluation of the Thermal Cross Sections of ^{239}Pu and ^{241}Pu," EPRI NP-1763, Electric Power Research Institute (1981).

[12]C. R. RICHEY, "EGGNIT: A Multigroup Cross Section Code," BNWL-1203, Pacific Northwest Laboratory (1969).

TABLE V

Thermal Neutron Cross-Section Data from Various Sources (Refs. 8 through 11)

Isotope	Neutrons per Fission, ν	Microscopic Fission Cross Section, σ_F(b)	Fission Non-1/v Factor, g_F	Microscopic Absorption Cross Section, σ_a (b)	Absorption Non-1/v Factor, g_a
^{238}Pu	2.90 ± 0.03	16.3 ± 0.5	0.95326 ± 0.01[a]	562.8 ± 20	0.95380 ± 0.01[a]
^{239}Pu	2.863 ± 0.010	754.84 ± 4.5	1.05353 ± 0.0015	1028.6 ± 5.1	1.07620 ± 0.00837
^{240}Pu	2.17 ± 0.01	0.058 ± 0.045	1.02511 ± 0.01[a]	290.06 ± 1.4	1.02639 ± 0.01[a]
^{241}Pu	2.918 ± 0.010	1003.8 ± 8	1.046 ± 0.006	1368.5 ± 10	1.037756 ± 0.006
^{242}Pu	---	---	---	18.485 ± 0.4	1.0100 ± 0.01[a]
Hydrogen	---	---	---	0.332 ± 0.002	1.000
Nitrogen	---	---	---	1.894 ± 0.05	1.000
Oxygen	---	---	---	(2.7 ± 0.25) × 10^{-4}	1.000

[a]Estimated uncertainty.

TABLE VI

Eta Calculation Parameters

Parameter	Value
Σ_{ax}	2.56843-2[a] ± 2.517-4 cm^{-1}
Σ_{at}	4.88332-2 ± 3.006-4 cm^{-1}
f	0.52596 ± 0.00609
F	0.99129 ± 0.00524
τ	26.564 ± 0.230 cm^2
L^2	3.5107 ± 0.0306 cm^2
Bg^2	2.4126-3 ± 2.265-4 cm^{-2}
$P(B)$	0.93188 ± 0.00602
$\bar{\eta}$	2.056 ± 0.037
η (2200 m/s)	2.100 ± 0.041

[a]Read as 2.56843 × 10^{-2}.

series of critical spheres containing $Pu(NO_3)_4$. The eigenvalues were plotted versus the hydrogen-to-plutonium (H/Pu) atomic ratio, and the curve that best fit the data was extrapolated to the curve for k_∞ of ($^{239}Pu + H_2O$) systems. This technique, attributed to Clark by Richey,[2] removes the calculational bias with varying H/Pu atomic ratio. This bias may be due to either calculated neutron leakage or plutonium concentration effects.

Eight critical systems in addition to the one reported here were employed in the extrapolation. These are the same eight systems evaluated by Richey[13] from experiments by Lloyd et al.[3] The k_{eff} of the spheres were computed using ENDF/B-IV cross-section data in an 18-group calculation with the HFN code.[14] Epithermal energy groups were obtained via application of the ETOG code[15] and the GAM portion of the EGGNIT code,[12] and the single thermal group data via the FLANGE code[16] followed by the TEMPEST portion of the EGGNIT code.

The calculated eigenvalues for these critical experiments using ENDF/B-IV data are shown in Table VII. Richey indicated the old data are best fitted to a straight line, which extrapolates to a minimum critical concentration between 7.1 and 7.2 g Pu/ℓ. The calculated curve shape is changed by both the addition of the present experiment and the recalculation of the older experiments using ENDF/B-IV library data. The best fit is obtained with a logarithmic function rather than a straight line. The eigenvalues from Table VII are plotted in Fig. 2, a semilog plot.

Although the eigenvalues in Table VII are functions of many variables, the only variable with a clear trend is the H/Pu atomic ratio. Such other variables as nitrate concentration, reflection, geometry, and ^{240}Pu content may well have associated biases, but

[13]C. R. RICHEY, *Nucl. Sci. Eng.*, **31**, 34 (1968).

[14]J. R. LILLEY, "Computer Code HFN–Multigroup, Multiregion Neutron Diffusion Theory in One Space Dimension," HW-71545, Hanford Atomic Products Operation (1961).

[15]D. E. KUSNER, R. A. DANNELS, and S. KELLMAN, "ETOG-1: A FORTRAN-IV Program to Process Data from the ENDF/B File to the MUFT, GAM and ANISN Formats," WCAP-3845-1 (ENDF-114), Westinghouse Electric Corporation (1969).

[16]H. C. HONECK and D. R. FINCH, "FLANGE-II (VERSION 71-1): A Code to Process Thermal Neutron Data from an ENDF/B Tape," DP-1278 (ENDF-152), Savannah River Laboratory (1971).

TABLE VII

Computed Eigenvalues for Critical Assemblies of Aqueous Solutions of $Pu(NO_3)_4$

H/Pu Atomic Ratio	Sphere Diameter (cm)	Reflector	Chemical Concentration (g/ℓ)			k_{eff} (Diffusion theory)
			Plutonium	NO_3	H_2O	
4.6 wt% ^{240}Pu						
754	35.6	Full water	33.0	162	924.9	1.0304
466	35.6	Full water	47.9	465	774.4	1.0382
354	29.2	Full water	73.0	86	970.9	1.0459
87	29.2	Full water	268.7	346	868.0	1.0531
1068	38.6	Full water	24.4	58	976.9	1.0251
553	38.6	Full water	38.7	517	736.6	1.0355
668	38.6	0.12-cm-thick stainless steel	39.0	64	977.8	1.0375
125	38.6	0.12-cm-thick stainless steel	172.8	483	765.8	1.0495
2.5 wt% ^{240}Pu						
2722.0	123.6	0.77-cm-thick aluminum	9.457	78.3	959.9	1.0196

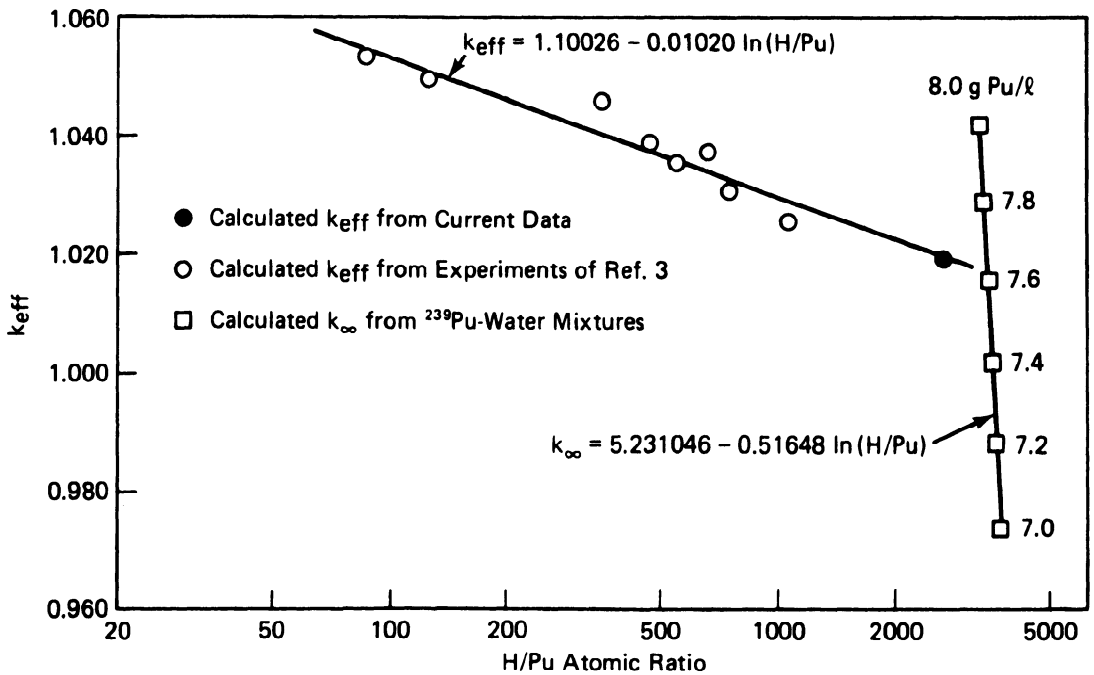

Fig. 2. Experimental eigenvalues and calculated ^{239}Pu-water data.

the limited data do not yield a significant bias function. Thus, the minimum critical concentration is determined using the assumption that all bias is a result of the H/Pu bias. Calculations were made to determine the infinite multiplication factor of plutonium-water mixtures and $Pu(NO_3)_4$-water systems. These results are shown in Table VIII. Figure 2 shows the calculated values of k_∞ plotted on the right side of the graph. The line through the experimental critical values is extrapolated until it intersects the k_∞ curve. This line was determined by a least-squares regression to fit an equation of the form

$$k_{eff} = a + b \ln(H/Pu) \quad . \quad (4)$$

The point of intersection yields the critical concentration with the calculational bias in k_{eff} as a function of the H/Pu atomic ratio eliminated. For plutonium-water mixtures, the minimum critical concentration is 7.59 g Pu/ℓ or H/Pu equal to 3495. The minimum critical concentration for $Pu(NO_3)_4$-water systems (Fig. 3) is 7.62 g Pu/ℓ or H/Pu of 3472.

The uncertainty in the plutonium concentration for the sphere experiment results in an uncertainty of less than ±0.1 g Pu/ℓ in the minimum critical concentration for aqueous plutonium systems. The impurity levels are so low that the inclusion or exclusion of impurities make <0.001 change in Δk of the sphere, which results in <0.01 g Pu/ℓ uncertainty in the minimum critical concentration.

TABLE VIII

Calculated k_∞ for ^{239}Pu-Water and $^{239}Pu(NO_3)_4$-Water

	Plutonium-Water		$Pu(NO_3)_4$-Water	
g Pu/ℓ	H/Pu	k_∞	H/Pu	k_∞
7.0	3789	0.9754	3782	0.9733
7.2	3683	0.9899	3676	0.9877
7.4	3584	1.0041	3577	1.0018
7.6	3489	1.0178	3482	1.0155
7.8	3400	1.0313	3393	1.0289
8.0	3315	1.0444	3308	1.0419

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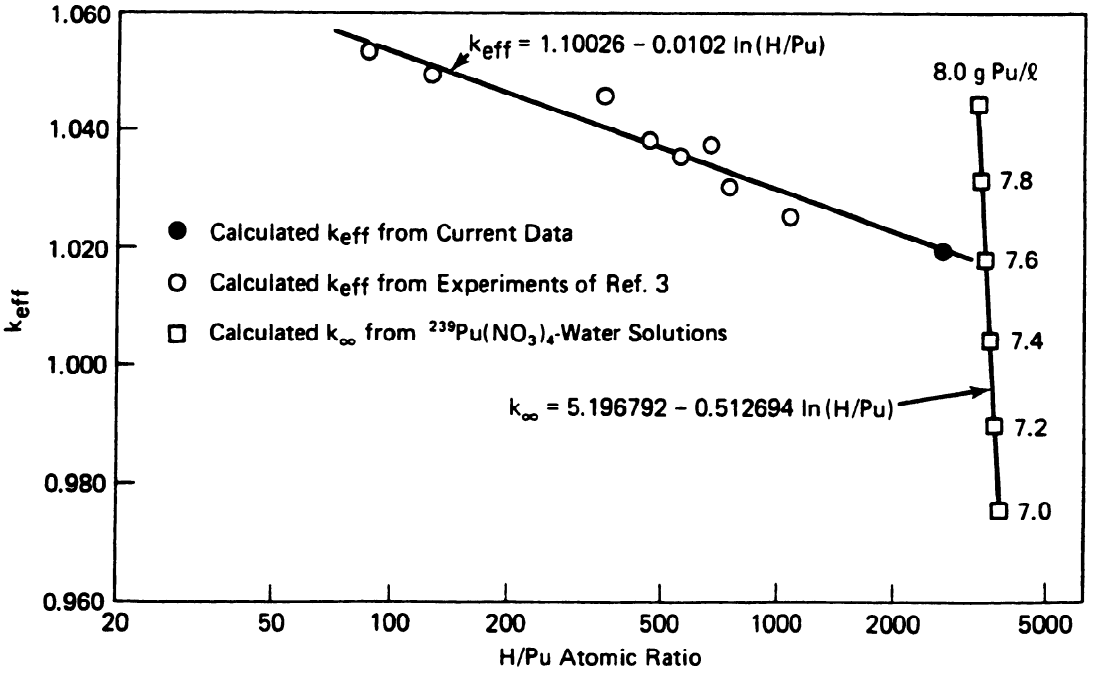

Fig. 3. Experimental eigenvalues and calculated $^{239}Pu(NO_3)_4$ data.