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LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA o LOS ALAMOS NEW MEXICO

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CRITICAL MASSES OF COMPOSITES OF Oy AND Pu-239-240 IN FLATTOP GEOMETRY

by

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ABSTRACT

Three critical plutonium-enriched uranium composites with plutonium of different Pu^{240} content are described. In these systems one gram of Pu^{240} is equivalent to ~ 0.63 grams of Pu^{239} and the analysis is presented which translates this datum to $\bar{\upsilon}(Pu^{240}, 2\ mev) = 3.32 \pm 0.14$ neutrons emitted per Pu^{240} fission induced by a 2 mev neutron.

I. INTRODUCTION

To provide information on the neutronic behaviour of the higher plutonium isotopes (specifically that of Pu^{240}), the critical mass of Oy(93.2% U^{235}), surrounding a 1.6 kg ball of alpha phase Pu(X% Pu^{239}) in Flattop geometry, has been measured for three values of X corresponding to three different plutonium irradiation histories. In view of the numerous previous measurements of critical configurations of Tu reflected Pu-Oy composites with low MWD/T plutonium, the present three critical assemblies yield only two new data points of a material substitution or perturbation character. Basically a substitution consists of interchanging a mass m(Pu) of Pu(X% Pu^{239}) for the same mass of Pu(X_o% Pu^{239}) and interchanging a mass Δm(Oy) of Oy(93.2% U^{235}) for a corresponding mass of Tu. As a substitution preserves uranium and plutonium densities, it essentially also preserves the collision mean free path distribution, and reactivity changes depend only on the neutron production cross section $\sigma_p = [\upsilon-1-\alpha]\sigma_f$. The reactivity balance equation is thus of the form

$$[\sigma_p(X)-\sigma_p(X_o)]m(Pu) = W[\sigma_p(Oy)-\sigma_p(Tu)]\Delta m(Oy) \quad (1)$$

where W is a statistical weight factor giving the importance of neutron production in the Oy-Tu substitution volume relative to that in the Pu ball. The quantity W, or its equivalent, must be deduced from a neutron transport computation for the critical assembly geometry involving the "known" nuclear properties of Pu^{239}, U^{235}, and U^{238}.

II. EXPERIMENTAL

The critical assembly machine used for this experiment is referred to as Flattop. It is a spherically symmetric geometry consisting of three parts:

a) A solid ball of the Pu isotopes at the center. (There are three balls, all 2.150" O.D. with 10 mil nickel coatings).

b) Three sets of interchangeable Oy-Tu shells that fit around the Pu balls. The two small sets are simple hemi-shells but one hemi-shell of the largest set is cut into a ring and cap.

c) A natural uranium (Tu) reflector, 19" O.D. and about 4-1/4" I.D.

Figure 1 shows a schematic of this assembly in horizontal cross-section. The Tu reflector is divided

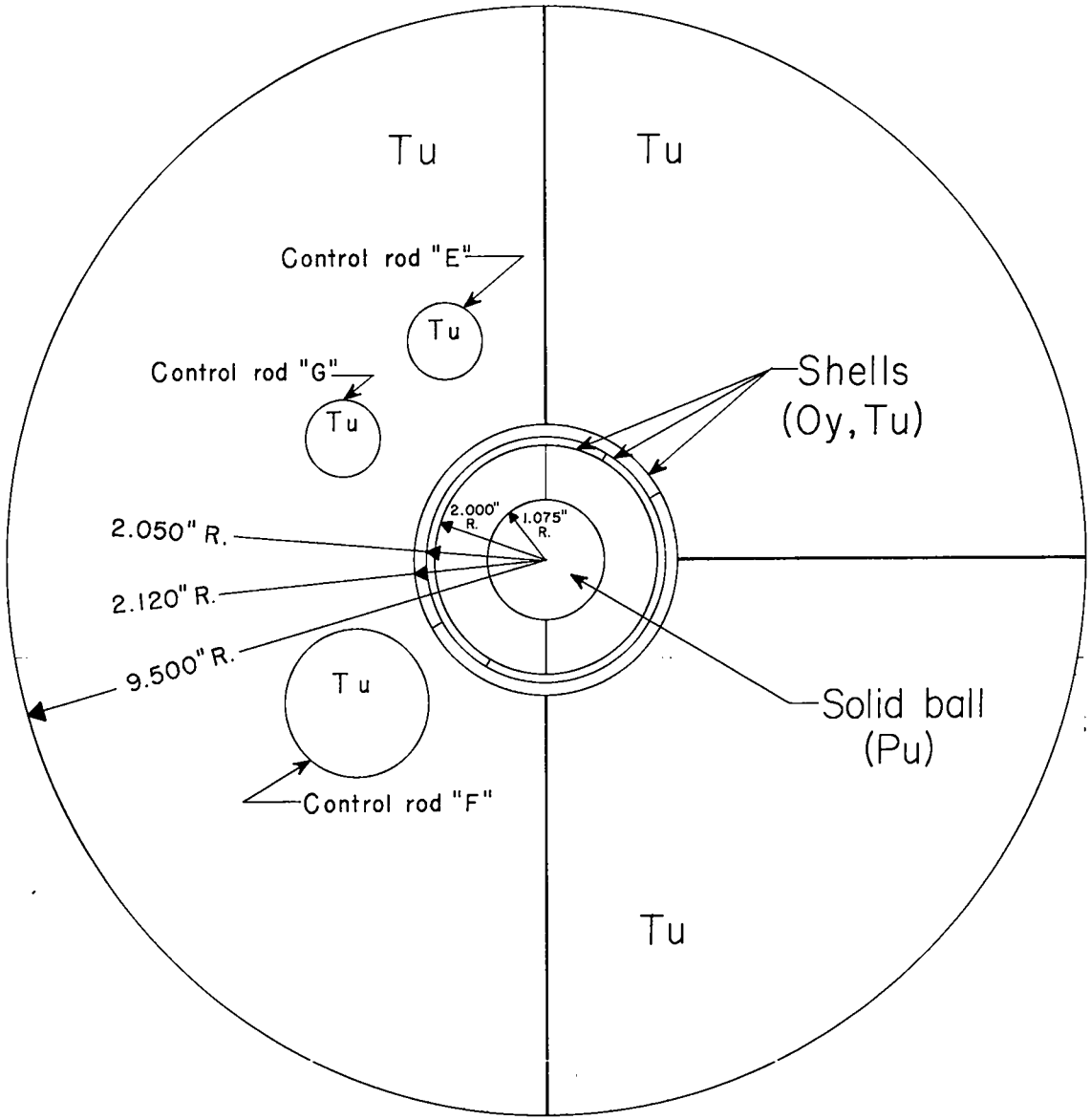

Fig. 1. Flattop Schematic

into a stationary hemisphere and two movable quarter spheres. There are three control rods of Tu in the stationary part of the reflector.

Table I gives the specifications of the three Pu cores. J-4591 has the lowest 240 content, J-4742 contains an intermediate amount of 240 and J-4744 has about 16% of this isotope. Part "A" of the table gives the mass in grams of Pu and Ni and atomic % of the various isotopes from mass spectrometer data.

Part "B", derived from the data of Part "A", gives the mass of each isotope present in a ball.

Approach to critical was observed by means of a reciprocal multiplication vs. mass plot. The data are plotted in Figure 2 as the mass of Oy replacing Tu in the assembly is increased. Table II shows the data in tabular form. Unit multiplication is defined with the Pu ball and small set of Oy shells in place (M_{Oy} = 8642 gms). Data points are obtained for each additional hemi-shell.

For the intermediate Pu core (J-4742) delayed critical was obtained with control rods fully inserted, both sets of small shells and the Oy ring of the large shell (M_{Oy} = 9755).

For the J-4591 core, a 7 minute period was measured when all rods were inserted with two sets of Oy shells and

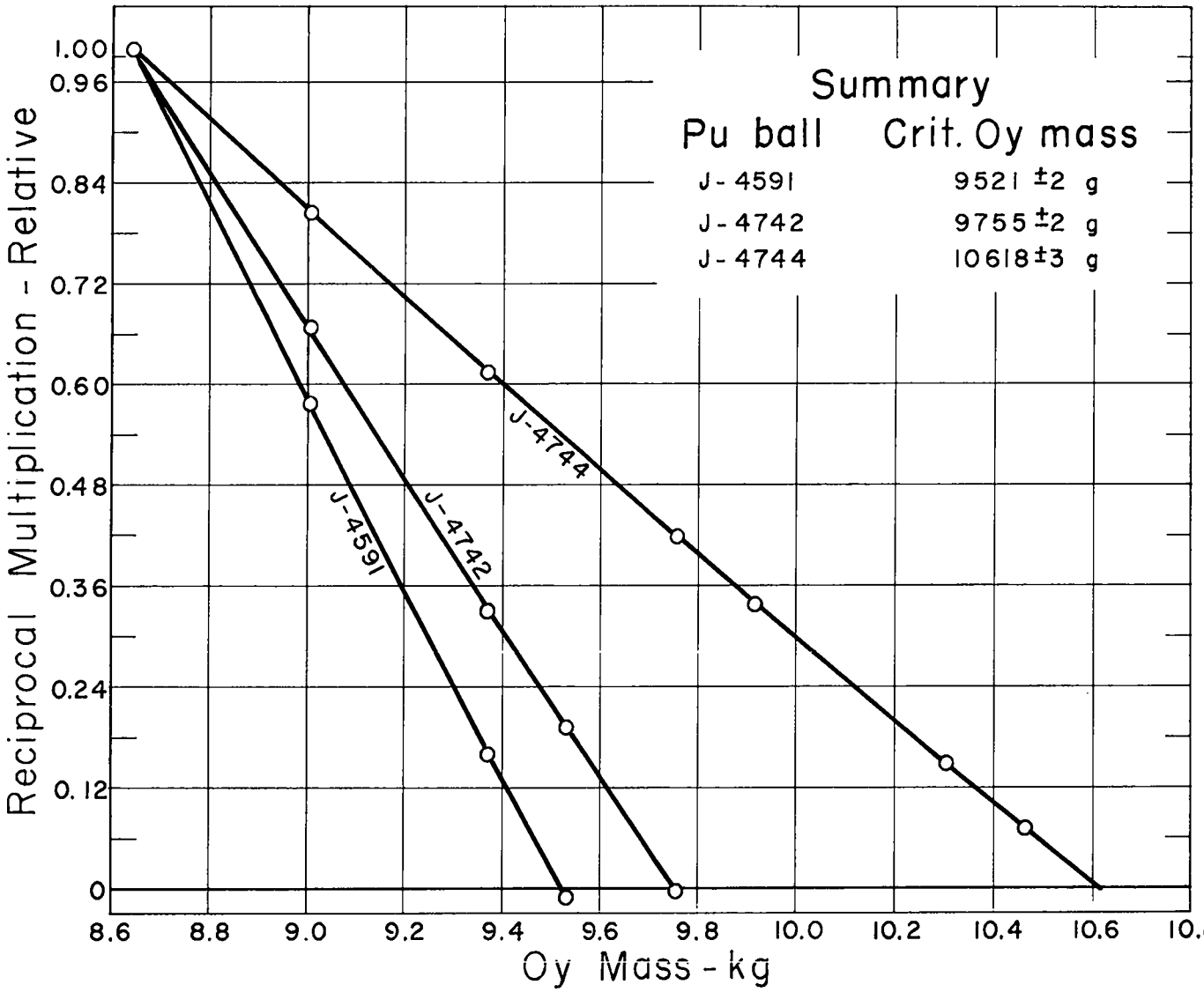

Fig. 2. Reciprocal Multiplication vs. Mass of Oy in the Pu-Oy Composite

the Oy cap. This period corresponds to a Δk of ~ 3.0 cents above delayed critical. By adding 160 grams Oy(an Oy cap) to the critical configuration of J-4742, a positive period of 4.7 sec. was obtained corresponding to ~ 53.8 cents above delayed critical and yielding the equivalence 2.96 g Oy $\equiv$ 1 cent. Delayed critical was thus estimated at 9530-(2.96 x 3) or 9521 gms Oy. This point is plotted on the graph as - 0.009 1/M corresponding to the Δk of 3 cents.

For the J-4744 core, a multiplication of 13.5 was observed with rods "in" and all the available Oy in the assembly. By extrapolation of the 1/M vs mass curve to zero 1/M, the critical mass of Oy was estimated as 10,618 $\pm$ 3 gms.

The measurements were repeated on successive days and the multiplications reproduced to $\pm$ 2% in all cases and on the average were about $\pm$ 1%.

III. ANALYTICAL

As the nickel-canned plutonium balls J-4591, J-4742, and J-4744 contain slightly different masses of plutonium and nickel, it is convenient to use the reactivity balance eq. (1) in the more general form

$$\Sigma \Delta k_i = \Sigma \frac{\delta k}{\delta m_i} \Delta m_i = 0 \qquad (2)$$

where Δm_i represents the change in mass of isotope i. The computation of the statistical weight factor W is now replaced by the computation of the average reactivity coefficients $(\overline{\delta k/\delta m_i})$ for i = Ni, Pu^{239}, U^{235}, and U^{238} in the substitutions J-4591 —> J-4742 and J-4742 —> J-4744.

Table III gives the results of nine DSN computations (in transport and S4 approximations with the sixteen group cross sections of N-2-753) on variants of the three Pu-Oy composite assemblies. Table IV gives the mass increments for the substitutions J-4591 —> J-4742 and J-4742 —> J-4744 (from Table I) together with the computed average reactivity coefficients (from Table III). As the multigroup specification of Pu^{240} given in N-2-753 is of questionable accuracy (specifically as regards the number of neutrons per fission), its role in the DSN computations may be considered as that of a hypothetical impurity which degrades the worth of plutonium thus permitting a larger Oy to Pu mass ratio in a critical geometry.

Letting k_i represent $(\overline{\delta k/\delta m_i})/(\overline{\delta k/\delta m_{239}})$, eq. (2) becomes, after introducing the mass increment and average reactivity coefficient values from Table IV

J-4591 —> J-4742: $38.53\ k_{240} + 3.21\ k_{241} = 27.67 \pm 0.25$ (2a)

J-4742 —> J-4744: $183.72\ k^1_{240} + 42.54\ k^1_{241} + 8.38\ k^1_{242} = 160.60 \pm 0.35$ (2b)

The uncertainties given on the right hand sides of eq. (2a) and (2b) originate from the measured increments $\Delta M(Oy)$ only: additional uncertainties originating in the plutonium isotopic analyses and computed reactivity coefficients will be remarked upon shortly. The prime label on k_{240} and k_{241} in eq. (2b) warns that the value of these coefficients may not be identical to the analogous coefficients in eq. (2a).

The coefficients k_{240}, k_{241}, etc., are primarily determined by the neutron production cross section $[\upsilon-1-\alpha]\sigma_f$. Fortunately, for the untangling of eqs. (2a) and (2b), the fission cross sections of the higher plutonium isotopes are recently known: $\sigma_f(Pu^{241})$ vs. energy (L. Smith, K. Smith, Henkel, and Thorpe, "Reports to AEC NCSAG, pp 33-4, Ap 28-29, 1960) is similar in shape and magnitude to that of the non-threshold fissioner Pu^{239} (BNL-325) whereas $\sigma_f(Pu^{242})$ vs energy (D. K. Butler, Phys. Rev. 117, 5, pp 1305-6, 1960) is similar in shape and magnitude to that of the threshold fissioner Pu^{239} (BNL-325). These data yield for a central Pu-Oy composite flux spectrum $\sigma_f(241) = 1.02\ \sigma_f(Pu^{239})$ and $\sigma_f(242) = 0.97\ \sigma_f(240)$, i.e., within experimental uncertainties in the cross section values, $\sigma_f(241) = \sigma_f(239)$ and $\sigma_f(242) = \sigma_f(240)$. In addition, the number of neutrons per thermal neutron induced fission of Pu^{241} has been measured and found to be $\upsilon_{th}(241) = 2.95\pm0.10$ (A.B. Smith, "Reports to AEC NCSAG, p 3, Sept. 19-21, 1960) thus comparing

closely with $\upsilon_{th}(239) = 2.89 \pm 0.03$ (BNL-325, 1st Sup.). These data, together with the empirical evidence that a) radiative capture cross-sections of the heavy elements are similar, and b) the numbers of neutrons per fission from isotopes of the same element are similar, lead to the following expectations: 1) $k_{241} = k^1_{241} \simeq [(\upsilon_{th}-1)\sigma_f(241)] / [(\upsilon_{th}-1)\sigma_f(239)] \simeq 1$, and 2) $k^1_{242} \simeq k^1_{240}$. Finally, because of the threshold character of $\sigma_f(240)$ and the known spectral softening accompanying the increase of Oy to Pu mass ratio in composite cores, one expects the worth of Pu^{240} relative to Pu^{239} to be less for the substitution J-4742 $\longrightarrow$ J-4744 than for the substitution J-4591 $\longrightarrow$ J-4742, i.e., $k^1_{240} < k_{240}$. The DSN computations listed in Table IV place this purely spectral effect (independent of the apriori poorly known value of $\upsilon(240)$) on the more quantitative basis, $k^1_{240} = [(118.5 \times 187.8) \div (120.4 \times 187.0)]\ k_{240} = 0.988\ k_{240}$.

With the assignments $k^1_{242} = k^1_{240} = 0.988\ k_{240}$ and $k^1_{241} = k_{241}$, eqs. (2a) and (2b) give $k_{240} = 0.642 \pm 0.011$ and $k_{241} = 0.91 \pm 0.05$. With the uncertainties originating in the plutonium isotopic analyses included, the above listed probable errors increase by about 40%, i.e., $k_{240} = 0.642 \pm 0.016$ and $k_{241} = 0.91 \pm 0.07$. One notes 1) this value of k_{241} is in reasonable agreement with the

expectation $k_{241} \simeq 1$ stated in the preceding paragraph, and 2) this value of k_{240} is in fortuitously good agreement with the value $[k_{240}]_{N-2-753} = 120.4/187.8 = 0.641$ obtained from Table IV as a consequence of the N-2-753 characterization of Pu^{240}. It is, of course, incorrect to use eqs. (2a) and (2b) in the above manner for the evaluation of both k_{240} and k_{241}: the experimental information on $\sigma_f(241)$ and $\upsilon(241)$ referenced earlier, and implying $k_{241} \simeq 1$, must be given some weight, thereby making it probable that $k_{241} > 0.91$. There is a practical advantage, at the present time, in choosing this ill-defined weight to yield $k_{241} = 1$ (permits consistent neutronics computations of plutonium systems on the convenient basis that the multigroup specifications of Pu^{239} and Pu^{240} are applicable to Pu^{241} and Pu^{242} respectively). With $k_{241} = 1$, eq. (2a) gives (including compositional uncertainties), $k_{240} = 0.635 \pm 0.009$ whereas eq. (2b) gives $k_{240} = 0.622 \pm 0.006$. From the weighted average $[k_{240}]_{Ave.} = 0.627 \pm 0.006$, one may conclude: "by reducing the magnitude of $\upsilon(240)$ listed in N-2-753 or N-2-731 by $1.5 \pm 0.7\%$, DSN computations using this revised listing will predict, on the basis of Pu^{239}-Pu^{241} and Pu^{240}-Pu^{242} cross section equivalences, the results of the two material substitution experiments within the uncertainties implied in the measurements of

critical mass and composition". This conclusion, useful for computational purposes, has little relevance for the question of how well do the material substitution measurements establish the value of $\upsilon(240)$. On the basis of uncertainties a) $\delta k_{241}/k_{241} = 10\%$, b) $\delta k_{242}/k_{242} = 20\%$, c) $\delta\sigma_f(240)/\sigma_f(240) = 5\%$, d) $\delta\alpha(240) = 50\%$ and e) those claimed in the critical mass and composition measurements, there is an implied uncertainty $\delta\upsilon(240)/\upsilon_{240} = 4.0\%$. There is also a probable bias in the N-2-753 characterization of the inelastic cross-sections of Pu^{239}: although this characterization yields good agreement with the measured Jezebel Rossi alpha spectral index, the predicted flux spectrum of Jezebel is softer than indicated by other data (leakage spectrum, U^{238}/U^{235} and Np^{237}/U^{235} fission cross-section ratios). If a different characterization of Pu^{239} inelastic scattering cross-sections is used which yields agreement with these latter Jezebel data but therefore yields disagreement with the Rossi alpha datum, call this characterization Z, the corresponding prediction for the Pu-Oy composites is $[k_{240}]_Z \tilde{=} 1.02\ [k_{240}]_{N-2-753}$ and the corresponding conclusion is to reduce the magnitude of $\upsilon(240)$ listed in N-2-753 by 3%. The 2% difference between $[k_{240}]_Z$ and $[k_{240}]_{N-2-753}$ is typical of the computational uncertainty, and one has $\upsilon(Pu^{240}) = (0.97 \pm 0.041)$ X

$[\upsilon(240)]_{N-2-753}$. This may be expressed explicitly for the case of fission induced by 2 mev neutrons (which corresponds approximately to the average energy of neutrons producing fission in Pu^{240} for metal plutonium systems):

$$\upsilon(Pu^{240},\ 2\ mev) = 3.32 \pm 0.14$$

With regard to the earlier conflicting estimates of $\upsilon(Pu^{240})$ from the replacement data of Topsy, Godiva, and Jezebel - υ(240, Topsy) = 3.88±0.20, υ(240, Godiva) = 3.26±0.24, and υ(240, Jezebel) = 3.27±0.17 (Engle, Hansen, Paxton, N.S.E. 8, 6, 1960) -, the present more accurate estimate points out the Topsy data as erroneous.

TABLE I

PLUTONIUM BALL MASS SPECIFICATIONS

A

	Mass (gms)		Isotopic Composition			
Ball	Pu	Ni	239	240	241	242
J-4591	1615.45	10.89	.9756	.0234	.0010	
J-4742	1610.30	11.76	.9497	.0473 ±.0005	.0030 ±.0001	
J-4744	1611.19	14.10	.8047	.161 ±.002	.0292 ±.0006	.0051±.0003

B

Ball	Mass (gms)				
	239	240	241	242	Ni
J-4591	1575.87	37.96	1.62		10.89
J-4742	1528.98	76.49±.8	4.83±.2		11.76
J-4744	1295.23	260.21±3.2	47.37±1.0	8.38±.5	14.10

TABLE II

1/M Vs. MASS Oy-DATA POINTS

M(Oy) gms \ Recip Mult	J-4591	J-4742	J-4744
8642	1.00	1.00	1.00
9006	0.578	0.669	0.806
9370	0.162	0.331	0.616
9530	τ = 7 min	0.193	
9755		0.000	0.420
9917			0.340
10302			0.149_5
10462			0.0740

TABLE III

SUMMARY OF DSN COMPUTATIONS FOR ESTABLISHING REACTIVITY COEFFICIENTS IN THE Pu-Oy COMPOSITES. MASS SPECIFICATIONS ARE IN GRAM UNITS.

Problem No.	Region I*		Region II	Region IV		Reproduction No. k
	$m(Pu^{239})$	$m(Pu^{240})$	m(Ni)	m(Oy)	m(Tu)	
1	1600.0	0	10	0	1138.9	1.012327
2	1620.0	0	10	0	1138.9	1.016083
3	1470.0	130.0	10	0	1138.9	1.003566
4	1470.0	130.0	15	0	1138.9	1.003700
5	1340.0	260.0	10	0	1138.9	0.994663
6	1600.0	0	10	562.7	569.4	1.020847
7	1470.0	130.0	10	562.7	569.4	1.012299
8	1470.0	130.0	10	1012.8	0	1.018449
9	1340.0	260.0	10	1125.3	0	1.012684

* Region I covers the radius limits 0-1.065 inches and represents the nominal volume of the alpha phase plutonium balls. Region II covers the radius limits 1.065-1.077 inches and represents the nominal volume occupied by the Nickel cladding. Region III covers the radius limits 1.077-2.052 inches and, as in the material replacement experiments, contains the fixed quantity of 9370 grams Oy(93.2% U^{235}). Region IV covers the radius limits 2.052-2.120 inches and corresponds to the nominal volume occupied by the Oy and Tu shells used for reactivity compensation. Region V, extending from 2.120 to 9.50 inches, contains Tu at 18.80 gms/cm^3 and corresponds to the nominal Flattop reflector.

TABLE IV

OBSERVED MASS INCREMENTS (GRAMS) AND COMPUTED REACTIVITY COEFFICIENTS FOR THE TWO MATERIAL SUBSTITUTION EXPERIMENTS

Substitution	Δm_{239}	Δm_{240}	Δm_{241}	Δm_{242}	Δm_{Ni}	Δm_{Oy} *
J-4591->J-4742	- 46.89	38.53	3.21	-	0.87	234±3
J-4742->J-4744	-233.75	183.72	42.54	8.38	2.34	864±3

	$\frac{\delta k}{\delta m_{239}}$ $(\times 10^6)$	$\frac{\delta k}{\delta m_{Ni}}$ $(\times 10^6)$	$\left(\frac{\delta k}{\delta m_{Oy}} - \frac{238}{235}\frac{\delta k}{\delta m_{Tu}}\right)$ $(\times 10^6)$	$\frac{\delta k}{\delta m_{240}}$ $(\times 10^6)$
J-4591->J-4742	187.8	26.8	15.33	120.4
J-4742->J-4744	187.0	26.8	15.76	118.5

* For each substitution, the increment Δm_{Oy} in Oy mass implies a decrement of $238\Delta m_{Oy}/235$ in Tu mass.