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MASTER

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**APRIL, MAY, JUNE, 1962**

**THE STAFFS OF  
NUCLEAR PHYSICS RESEARCH OPERATION  
and  
APPLIED PHYSICS OPERATION**

JULY 16, 1962

**HANFORD LABORATORIES**

HANFORD ATOMIC PRODUCTS OPERATION  
RICHLAND, WASHINGTON

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EXPERIMENTAL NUCLEAR PHYSICSTests of a New Ionization-Density Discriminator

D. G. Foster, Jr.

Circuits designed to distinguish charged particles of different mass by the difference in time distribution of the light which they produce in a scintillator<sup>(1-3)</sup> have been used widely in recent years. One of the most important applications has been in distinguishing between recoil electrons and recoil protons, due to incident photons and neutrons respectively, in a hydrogenous scintillator.

We describe here performance tests on a circuit designed by J. T. Russell\* specifically for use with the Hanford Laboratories fast neutron time-of-flight facility. The evaluation of a circuit for any particular application includes both its compatibility with the remainder of the equipment and its performance characteristics per se. In the present instance, it is desirable to have a circuit which requires only one signal from the photomultiplier, and on which any critical adjustments can be made in the control room approximately 75 feet from the detector. Many of the best circuits designed previously make use of pulses from both the anode and the last dynode of the photomultiplier, and the tube is operated in a region of response linear with respect to input light intensity. The existing Hanford Laboratories time-of-flight apparatus depends on the use of a multiplier operated well into saturation, and determinations of pulse height and pulse shape must be made from a dynode several steps removed from the anode. The restriction on the location of the circuit itself is imposed by the personnel radiation hazard near the detector.

Since the circuit has not been described previously, a brief discussion of it will be included here. The arrangement is shown schematically in Figure 1.

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\* Instrument Research and Development Operation

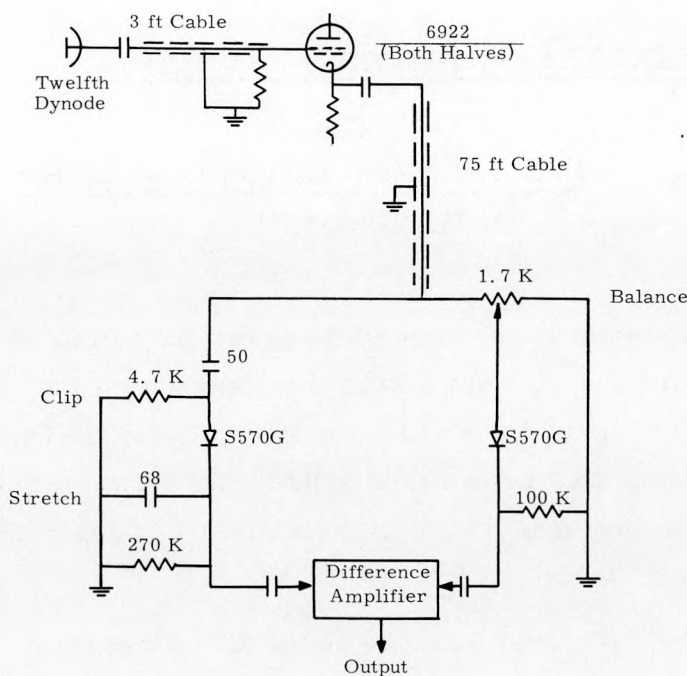


FIGURE 1

Essential Parts of the Ionization-density Discriminator

The short charge pulse due to the detection of an event in the fast organic scintillator by a 14-stage photomultiplier is dumped onto a 15-micro-second integrating time constant. This signal, with its slowly-changing tail arising from the slow components which are of interest, is transmitted to the cable by a cathode follower. The cable is unterminated, but the succeeding circuits are too slow to be disturbed by the reflections.

At the input to the discriminator, the signal is split into two channels. In the left-hand channel it is differentiated, and stretched back to its original time constant by a high-speed S570G diode. In the right-hand channel the signal is attenuated, and then fed through a diode of the same type. The purpose of the second diode is to compensate, to first order, for the non-linearity of the stretching diode for small signals; for large signals the pulse is unaltered by the diode.

The two channels are then fed to a differential pulse amplifier,\* which amplifies the difference between the unaltered pulse in the right-hand channel and the initial height of the pulse as given by the stretched pulse in the left-hand channel. The attenuator in the right-hand channel is adjusted until the amplified difference signal arising from a recoil electron never goes positive. The slow "tail" on the pulse from a recoil proton then goes substantially positive, and serves as an indication that the event was due originally to a neutron.

The best method<sup>(3)</sup> of testing a circuit of this type is to determine the number of output pulses as a function of both input and output pulse height, using a two-dimensional pulse height analyzer. Since such an analyzer is not available, a one-dimensional multichannel analyzer in coincidence with a single channel analyzer is used. The type of two-dimensional spectrum to be expected from a source producing both neutrons and photons is diagrammed in Figure 2, and the measurements are equivalent to obtaining selected vertical strips from this plot.

In Figure 2, the height of the original pulse is plotted on the horizontal axis, and the height of the output pulse from the shape discriminator circuit is plotted vertically. The horizontal coordinate is a nonlinear but monotonic function of particle energy also. Electron and proton recoils form two families of pulses which fuse together at low (and frequently at high) energies. Figure 3 shows contours taken along the lines of constant input pulse-height labelled A and B in Figure 2. From these plots it is evident that the two families are clearly separated at moderate energies, but not at low energies.

For electronic simplicity, the electron recoils are normally eliminated by setting a simple discriminator threshold, at the output of the shape discriminator, to reject a specified fraction of the electron recoils

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\* The first interstage section of a commercial nonoverloading linear amplifier of the Chase-Higginbotham type (Technical Measurement Corporation, Type AL-4A). An additional attenuator is required to prevent saturation of the remaining stages.

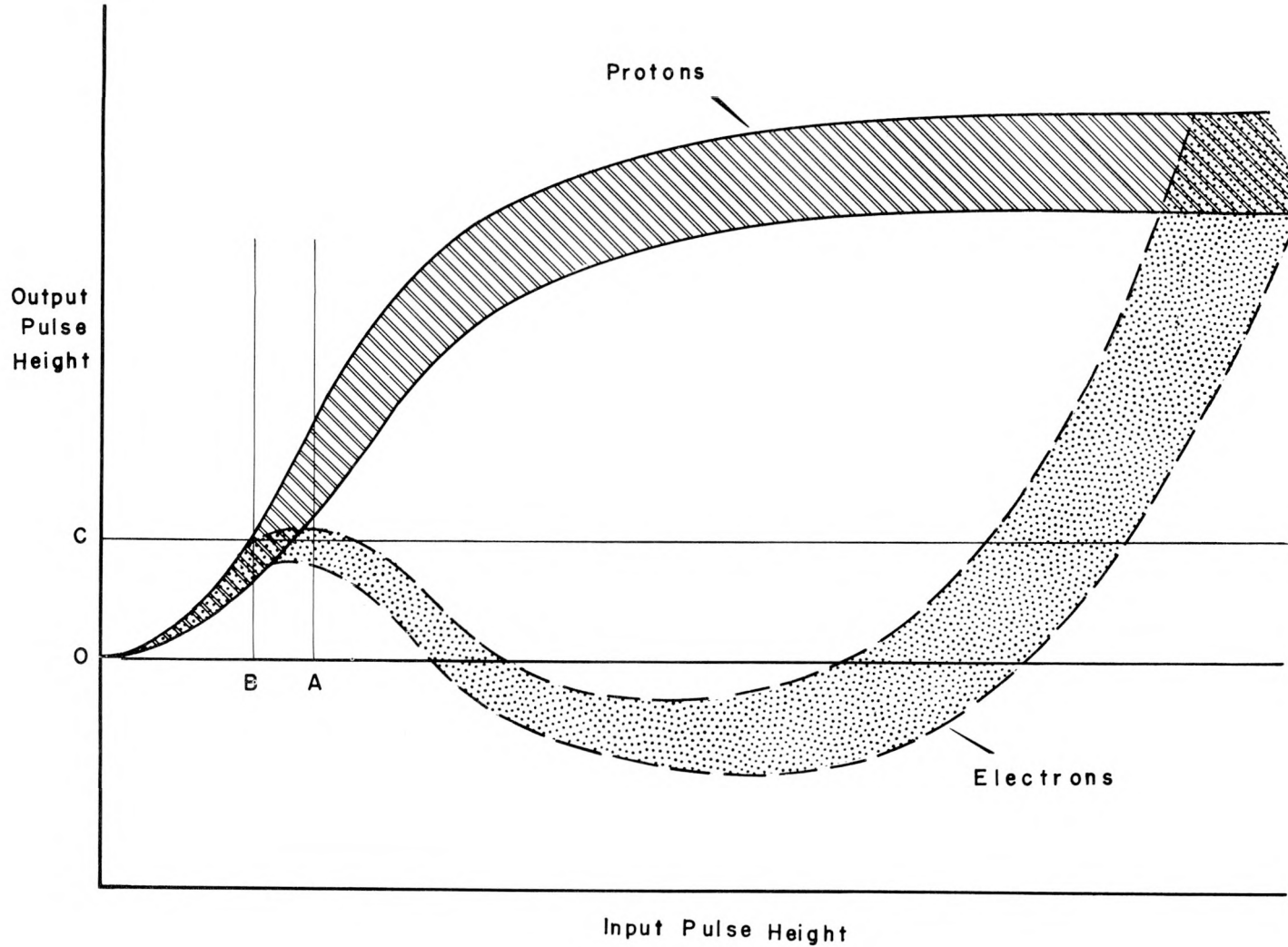


FIGURE 2

Typical Operating Characteristics of the Discriminator (schematic). The Abscissa may be Interpreted as Particle Energy, but the Scale is Nonlinear and Different for Electrons and Protons. The Rapid Rise in Output Pulse Height for Electrons with Large Input Pulse Heights Occurs Typically at an Electron Energy of the Order of 4 Mev.

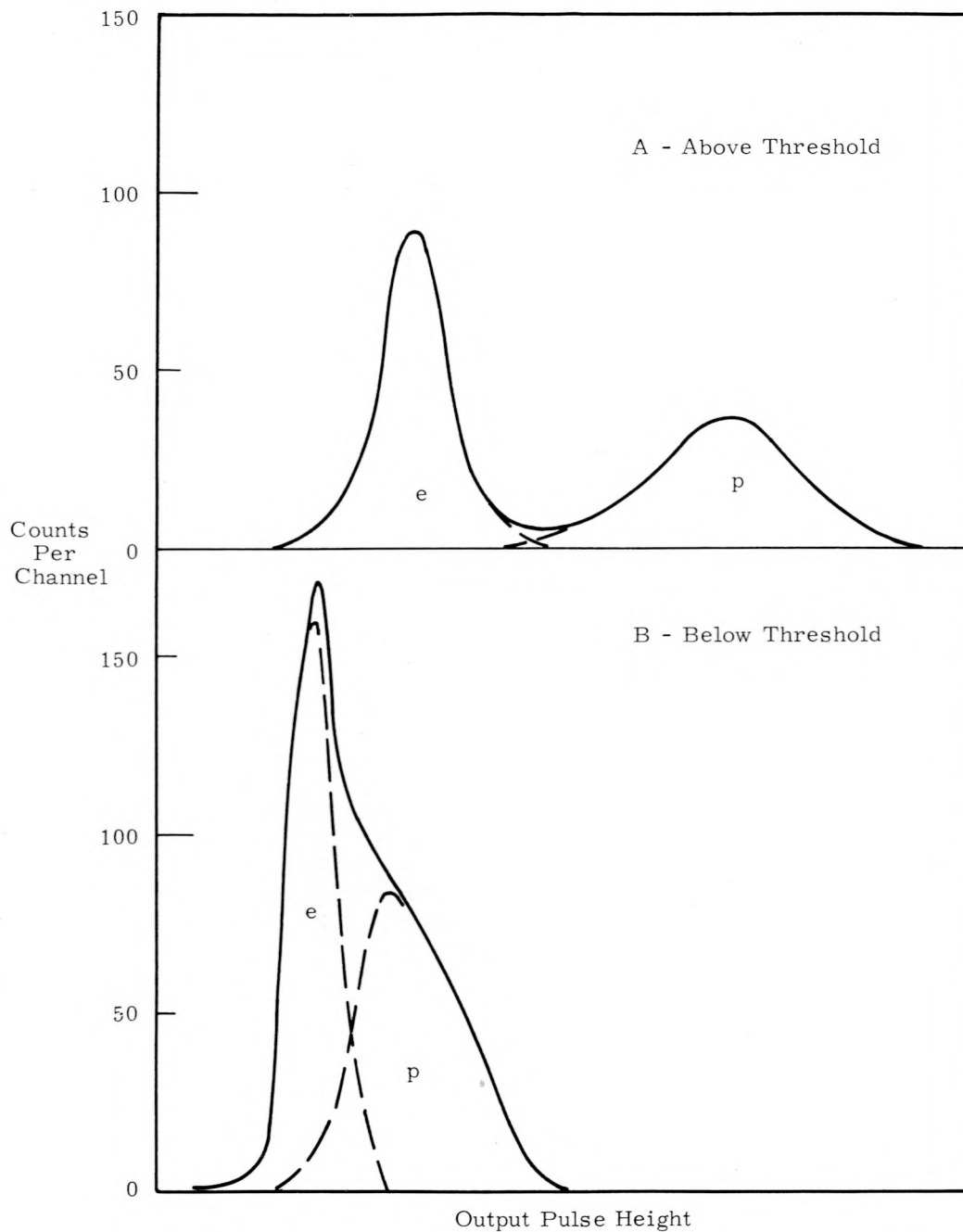


FIGURE 3

Output Pulse Height Spectra Taken Along the Lines of Constant Input Pulse Height Marked in Figure 2. These Data were Taken with a Stilbene Scintillator Mounted on a 6810A Photomultiplier. Line A is Taken at an Input Pulse Height Corresponding to Recoil Electron Energies Near 130 kev. Line B Corresponds to Electron Energies Near 60 kev.

irrespective of energy. In Figure 2 such a threshold is represented by the Line C. Figure 4 shows a plot of the fraction of proton recoils whose output pulse heights lie above this threshold. The steepness of this plot is a measure of the resolution of the circuit and the position of the cutoff may be defined as the threshold of the circuit.

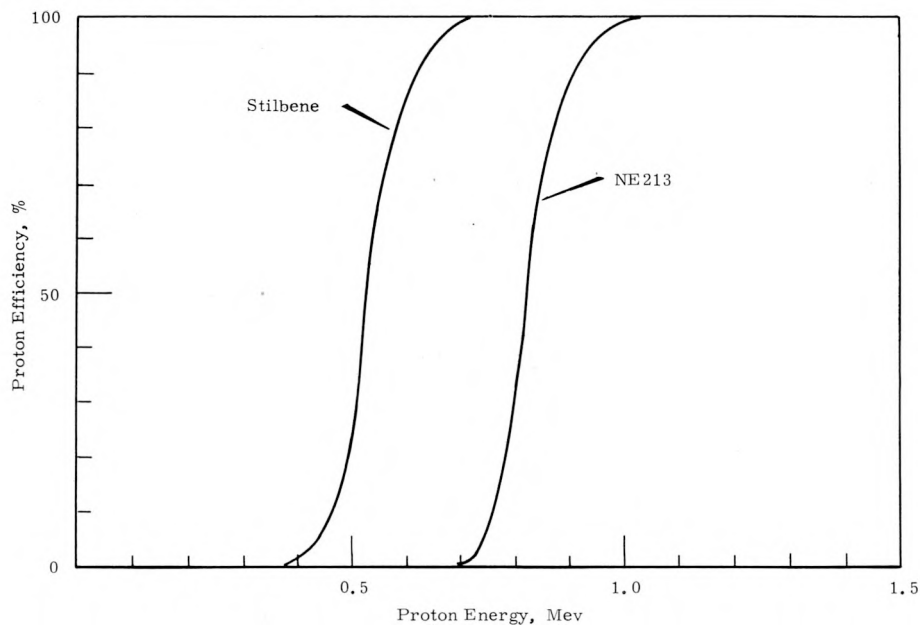


FIGURE 4

Detection Efficiency as a Function of Energy for Recoil Protons Which Produce Output Pulse Heights Greater than the Discriminator Setting Shown as Line C in Figure 2 (chosen to reject 99 percent of all recoil electrons in a representative case).

The particular data shown in Figure 4 are for an electron threshold (line C in Figure 2) chosen to reject 99 percent of all the pulses produced by a natural thorium gamma source, which is taken as being approximately representative of the backgrounds encountered in time-of-flight service. Two curves are shown, one for a transstilbene scintillator mounted on an RCA 6810A, and the other for a liquid scintillator\* (NE213) mounted on an Amperex 56 AVP photomultiplier. The abscissa is marked in units of proton

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\* Nuclear Enterprises Ltd., 1750 Pembina Highway, Winnipeg 9, Manitoba, Canada. The cell containing the liquid is 2 inches in diameter and 2 inches long, and is made of stainless steel lined with an aluminum foil reflector.

energy. The ratio of the light intensity produced by protons to that produced by electrons differs markedly for the two scintillators. The plots are based on the ratios measured by Borek and Anderson<sup>(4)</sup> for stilbene, and by Batchelor et al.<sup>(5)</sup> for NE213. The threshold (taken to be the energy at which the efficiency falls to 50 percent) is thus seen to be 0.53 Mev for stilbene and 0.82 Mev for NE213.

These thresholds are determined principally by the nonlinearity of the stretching diode for small pulses, and the lowest threshold is obtained by using the largest input signal. Large light signals into the photomultiplier, however, will saturate the tube and distort the shape of the pulse. This saturation accounts for the fusing of the electron and proton families at high energies (Figure 2).

Saturation can be prevented by keeping the input pulse height as small as possible. The best compromise between threshold and saturation occurs for signals taken from the twelfth dynode in both the 6810A and 56 AVP phototubes. It is worth noting, however, that not all experimental situations lead to large numbers of energetic recoil electrons, so that the saturation may be entirely tolerable. On the other hand, it has been found that minimum-ionizing cosmic ray mesons passing through a 5-inch diameter liquid scintillation counter (mounted on a 58 AVP photomultiplier), in which they can leave up to 15 Mev (equivalent to 20 Mev for protons), cannot be suppressed with this circuit.

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EXPERIMENTAL REACTOR PHYSICS

Lutetium as a Spectral Index Detector - Part III\*

L. C. Schmid and W. P. Stinson

The activities produced in lutetium foils are a function of the thickness of the foil because of self shielding by a resonance in  $\text{Lu}^{176}$  at 0.142 ev and other resonances at higher energies in  $\text{Lu}^{175}$ . Originally foils 9.7  $\text{mg}/\text{cm}^2$  thick were chosen for the lutetium calibration experiments. <sup>(1)</sup> This thickness was chosen to allow sufficient count rates and still not appreciably shield the resonance at 0.142 ev in  $\text{Lu}^{176}$ . In order to choose this optimum thickness, lutetium foils of various thicknesses were irradiated in the Thermal Test Reactor (TTR). <sup>(2)</sup> Some of the foils were covered with 0.040-inch thick cadmium to obtain the variation of epithermal neutron activations as a function of thickness of the lutetium foils. Since these measurements were made, Jacks <sup>(3)</sup> has reported results of similar measurements. The purpose of this report is to compare results and present a set of data for correcting lutetium activity measurements from thick foils to that for thin foils.

In order to make foils of various thicknesses, slurries which contained different concentrations of  $\text{Lu}_2\text{O}_3$  powder and water were allowed to evaporate in a disc-shaped aluminum capsule, 1/2-inch in diameter. The thin layers of  $\text{Lu}_2\text{O}_3$  were then covered by more aluminum to completely enclose the  $\text{Lu}_2\text{O}_3$ . The weights of  $\text{Lu}_2\text{O}_3$  were determined by weighing the aluminum capsule before and after the  $\text{Lu}_2\text{O}_3$  had been dried in it.

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\* Part I. "Experimental Technique," Nuclear Physics Research Quarterly Report - January, February, March, 1960. HW-64866. April, 1960.

Part II. "Resonance Integrals," Physics Research Quarterly Report - January, February, March, 1961. HW-69475. April, 1961.

The foils were irradiated on the rotator of the TTR and the  $\text{Lu}^{176\text{m}}$  and  $\text{Lu}^{177}$  gamma-ray activities were separated by counting the foils at different times. <sup>(1)</sup> In addition to the  $\text{Lu}_2\text{O}_3$  foils, "blank" foils containing only aluminum and evaporated water were irradiated and counted along with the other foils. The blank foils were used to monitor the activity induced in the aluminum capsules. This activity was from manganese which was present in the aluminum as an impurity since the foils were counted after the aluminum activity had decayed.

Foils which varied in thickness from  $0.335 \text{ mg/cm}^2$  to  $156 \text{ mg/cm}^2$  of  $\text{Lu}_2\text{O}_3$  and which were not surrounded by cadmium were irradiated. The activity from  $\text{Lu}^{177}$  and from  $\text{Lu}^{176\text{m}}$  was measured and the results are shown in Figure 1. In order to demonstrate the self-shielding effects on the induced activities, specific activities (i. e., counts/unit weight) are plotted versus the thickness in milligrams per square centimeter. For no shielding the specific activities would be constant. The errors shown in Figure 1 are those due only to counting statistics. The fluctuations of the data points about the smooth curve are caused mainly by the errors on the weights which could be as much as  $0.05 \text{ mg/cm}^2$ .

Foils which varied in thickness from  $0.50 \text{ mg/cm}^2$  to  $32 \text{ mg/cm}^2$  of  $\text{Lu}_2\text{O}_3$  and which were covered with 0.040-inch cadmium were also irradiated. From these foils the activations  $A_{\text{ec}}$  due to neutrons which were able to penetrate the cadmium were determined. The results are shown in Figure 2 as the ratio of the epicadmium activations  $A_{\text{ec}}$  and the activations  $A_{\text{sc}}$ . The activity  $A_{\text{sc}}$  is the difference between the activity from the uncovered foil and that from the covered foil. The ratios were determined by taking values from curves which were visual best fits to the data points. Cadmium ratios were calculated from the values and the ratio  $\frac{A_{\text{ec}}}{A_{\text{sc}}}$  is the reciprocal of one less than the cadmium ratio. Similar curves from Reference 3 are shown in Figure 2 also.

The data for  $\text{Lu}^{177}$  do not agree with that from Reference 3 for thicknesses less than  $28 \text{ mg/cm}^2$ . We did not observe an increase in cadmium ratio with thicknesses up to  $28 \text{ mg/cm}^2$ . The rate of increase in the curves

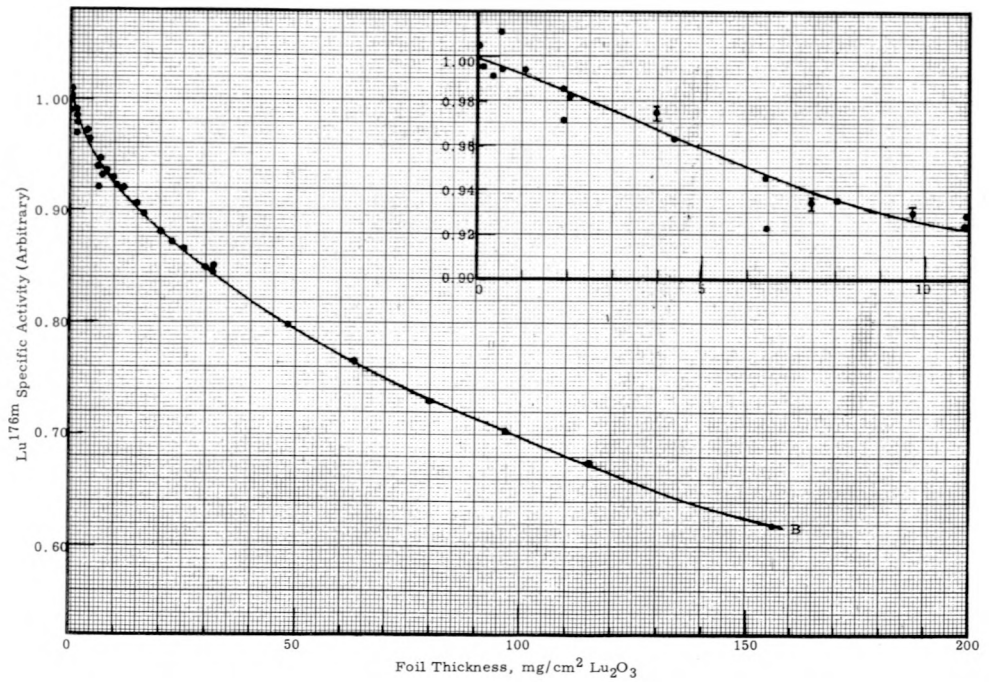
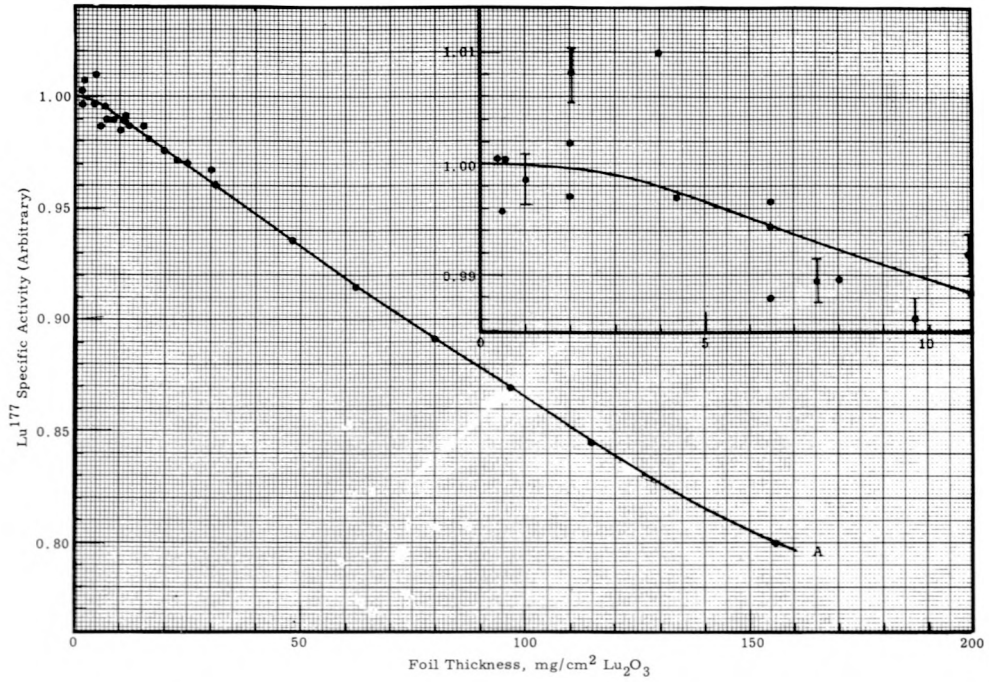


FIGURE 1  
Relative Specific Activities for Lu<sup>177</sup> and Lu<sup>176m</sup>

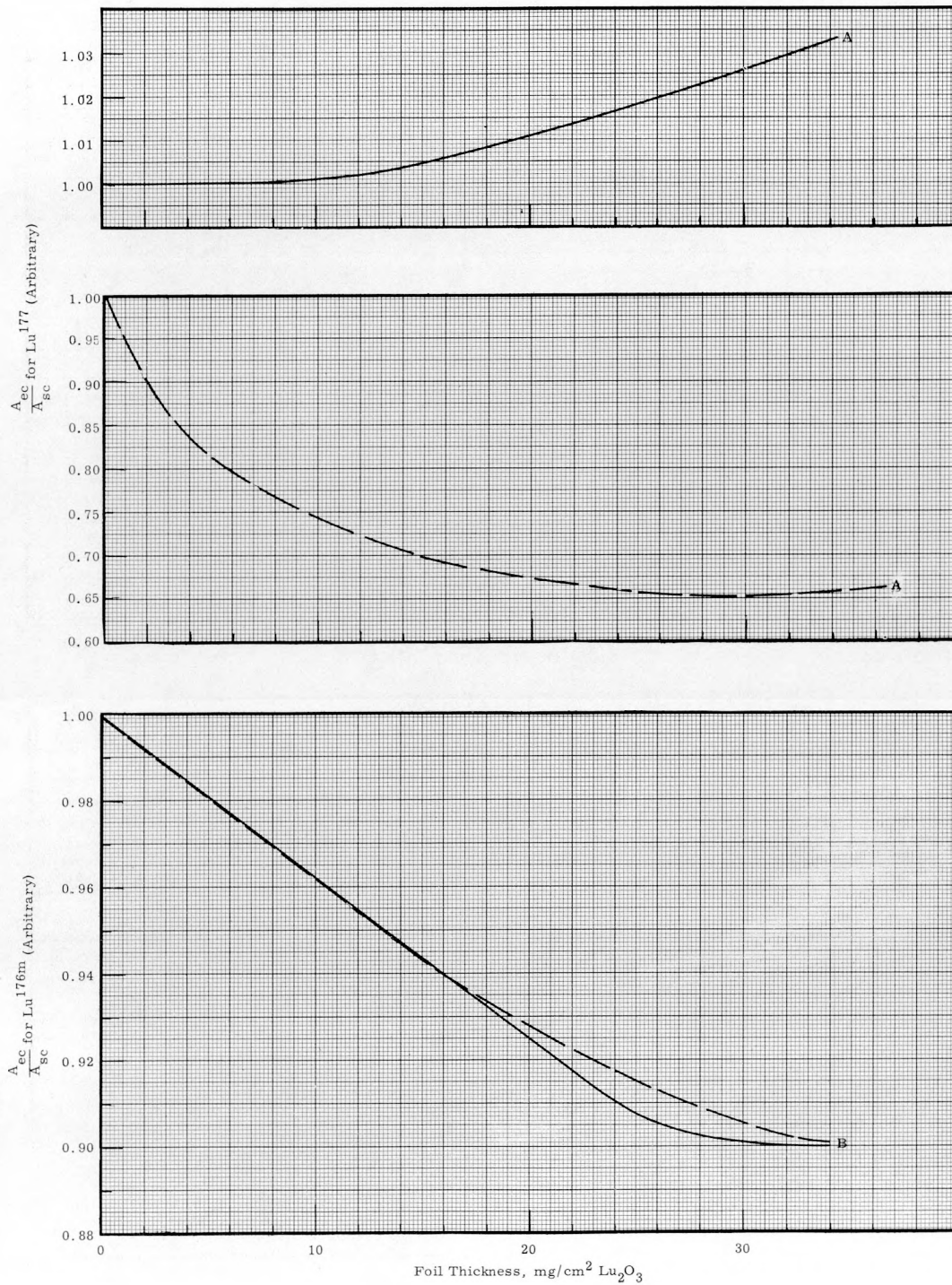


FIGURE 2

Ratio of Epicadmium to Subcadmium Activities  
In both cases, the dashed curves are taken from Reference 3.

for thickness between  $28 \text{ mg/cm}^2$  and  $34 \text{ mg/cm}^2$  was the same. The activity  $A_{ec}$  per unit weight remained constant for  $\text{Lu}^{177}$ . Therefore the change in  $\frac{A_{ec}}{A_{sc}}$  for  $\text{Lu}^{177}$  as a function of increasing thickness of the lutetium foils is due to the decrease in activity in  $A_{sc}$  per unit weight because of the self-shielding of the 0.142 ev resonance in the  $\text{Lu}^{176}$  cross-section.

The data agree with that from Reference 3 to within 1 percent for  $\text{Lu}^{176m}$ . The change in  $\frac{A_{ec}}{A_{sc}}$  for  $\text{Lu}^{176m}$  as a function of increasing thickness of the lutetium foils is due mainly to resonances, in the epithermal energy region, which reduces the activities  $A_{ec}$  per unit weight as the thickness of the foils is increased. The effect of this reduction on the ratio  $\frac{A_{ec}}{A_{sc}}$  for  $\text{Lu}^{177}$  is partially compensated for by a reduction in the activity  $A_{sc}$  because of the self-shielding effect of the 0.142-ev resonance in  $\text{Lu}^{176}$ .

As a result of the shielding by the 0.142 ev resonance in  $\text{Lu}^{176}$  the ratio  $\frac{A_{sc}^{177}}{A_{sc}^{176m}}$  is a constant for the thicknesses which were investigated (up to  $32 \text{ mg/cm}^2$ ). Thus, spectral index measurements which use cadmium ratio measurements to calculate  $\frac{A_{sc}^{177}}{A_{sc}^{176m}}$  should be independent of the thickness of the lutetium foil for this thickness range.

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Subcritical Measurements with 1.8 wt % Pu-Al Rods in Light Water

W. P. Stinson and L. C. Schmid

Subcritical experiments with 1.82 wt % Pu-Al rods in light water have been conducted as part of a series of experiments which utilize plutonium of various concentrations of the isotope  $\text{Pu}^{240}$ . The purpose of the experiments is twofold. First, the experiments furnish critical-mass data which allow more accurate nuclear safety specifications to be set for the reprocessing of plutonium fuels. Secondly, the data can be used as check points in the calculation of light-water moderated lattices which contain plutonium.

Neutron multiplication and exponential experiments were conducted in the TTR reactor room using a tank 4 feet in diameter and 5 feet deep.<sup>(1, 2)</sup> The rods of 1.8 wt % Pu-Al alloy were those fabricated for the PRTR.<sup>(3)</sup> They were subsequently cut into halves and resealed to form rods 44 inches in length. These rods were 0.500 inches in diameter and were sealed in Zircaloy-2 tubes with a 0.030-inch thick wall and 0.566 inches in outside diameter. One end of the rod contained helium in a 0.400-inch long region between the alloy and the end cap. The end caps were of various thicknesses and shapes.

The weights of the individual end caps and the section of tubing beyond the Pu-Al alloy varied from 11.0 to 14.0 g per rod with an average of 12.4 g per rod for the end next to the neutron source and varied from 14.5 to 27.0 g per rod with an average of 21.6 g per rod for the opposite end. An exploded view of a rod with a sample of each type of end cap is shown in Figure 1.

The average plutonium content was 7.12 g per rod. The amount of the  $\text{Pu}^{240}$  isotope in the plutonium varied but was principally 5 percent or 6 percent as shown in Table I. The distribution of the  $\text{Pu}^{240}$  fuel rods was random throughout the lattice. An average composition for the plutonium by weight is 93.97  $\text{Pu}^{239}$ , 5.58  $\text{Pu}^{240}$ , 0.44  $\text{Pu}^{241}$ , and 0.014  $\text{Pu}^{242}$ . An average composition for the aluminum by weight is 0.69 silicon, 1.61 nickel, 0.49 iron and 97.21 aluminum.

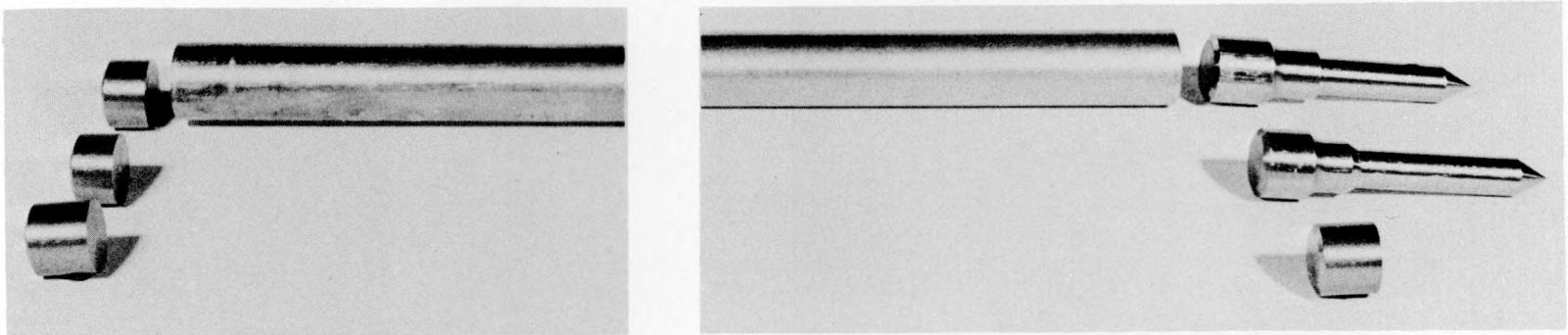


FIGURE 1  
A Fuel Element Rod with Typical End Caps

TABLE I

PLUTONIUM AND ZIRCALOY CONTENT OF THE Pu-Al RODS

<u>Number of Rods</u>	<u>Original Pu Content Per Rod, g</u>	<u>Pu<sup>240</sup>, %</u>	<u>Zircaloy, Grams Per Rod*</u>	<u>Al Per Rod, g</u>	<u>wt % Pu in Al</u>
211	7.06	5.05	234.9	382.0	1.807
259	7.19	6.00	238.3	382.3	1.840
27	7.00	5.58	234.9	382.1	1.807
497 Total	7.12**	5.58**	236.7**	382.2**	1.824**

\* Does not include the Zircaloy in end caps or tubes beyond the end of the fuel rod core.

\*\* Average

The lattices were hexagonal in shape and placed so as to provide effectively infinite water reflection on all sides. The lattice plates were constructed from lucite except for the top one which was made of aluminum 1/4-inch thick and which was positioned 2 inches above the end of the Pu-Al alloy. There were five sets of lattice plates used. The center-to-center spacing of the fuel element positions for the sets were 0.75-in., 0.80-in., 0.85-in., 0.90-in., and 0.95-in. Each rod was enclosed in a lucite tube, which was 0.650 inches in outside diameter and which had a wall thickness of 0.032 inches, before positioning it in the lattice. Water was allowed to occupy the spaces between the rod and the lucite tube in order to minimize voids. Previously, it has been determined that the lucite tubes do not affect the results. <sup>(4)</sup>

The experiments were conducted using critical approach and exponential measurement techniques. It was necessary to have control and safety rod thimbles in the loading during the critical approach. Therefore, once the loading contained 96 percent of the critical mass, the thimbles were removed. § This "back-off" technique resulted in a more accurate measurement than the critical approach since all the poison from the safety and control rod channels was removed. An external neutron source of plutonium and beryllium containing 80 grams of plutonium was positioned on

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§ From the system and multiplication data were obtained as fuel rods were removed.

the longitudinal axis and 4 inches from one end of the cylindrical array. Four  $\text{BF}_3$  proportional counters were used to detect the neutrons and were placed adjacent to the end of the array opposite the neutron source. One counter was positioned in the center and the other three were positioned symmetrically about the center and on the outside edge of the lattice when 1/4 of the critical mass had been loaded.

The critical number of rods was obtained by fitting a straight line, by the method of least squares, to  $N/\phi$  plotted against  $N$ . The  $N$  is the number of rods in the system and  $\phi$  is the flux as measured by the  $\text{BF}_3$  counter. The quantity  $N/\phi$  was used for the plots instead of the conventional  $1/\phi$  in order to remove the effect of the increasing background of neutrons from the  $(\alpha - n)$  reaction in the Pu-Al as more rods were added. The plots of  $N/\phi$  were linear between 60 percent and 96 percent of the critical mass. The exponential measurements were made when the lattice contained approximately 60 percent of its critical mass. Vertical transverses were taken along the central axis of the loadings with a  $\text{BF}_3$  counter. These data were plotted on semilog paper and a straight line was fitted to a plot of  $\ln \phi$  versus the position along the axis. The slope  $\gamma$  of this straight line was used in the material buckling equation

$$B_m^2 = \left( \frac{j_0}{R_e + \lambda} \right)^2 - \gamma^2$$

The  $R_e$  is the radius of the equivalent bare cylindrical loading (i. e.,

$R_e = \ell \sqrt{\frac{N_e \sqrt{3}}{2\pi}}$  where  $\ell$  is the lattice spacing and  $N_e$  is the number of rods in the exponential loading). The extrapolation distance plus reflector savings  $\lambda$  was assumed to be the same vertically and horizontally. Thus, the critical buckling is

$$B_c^2 = \left( \frac{j_0}{R_c + \lambda} \right)^2 + \left( \frac{\pi}{H + 2\lambda} \right)^2 = B_m^2$$

where  $R_c = \ell \sqrt{\frac{N_c \sqrt{3}}{2\pi}}$  and H is the length of the Pu-Al alloy. By equating the material and critical bucklings, values of  $\lambda$  were calculated for each lattice.

The data obtained for the five lattices are summarized in Table II. The critical mass and buckling data are presented in Figure 2. The value of  $\lambda$  for an H/Pu ratio of 1204 does not form a consistent curve with the other values for  $\lambda$ . The value of  $\lambda$  is high and the corresponding buckling is low because of an inconsistent value for  $\gamma$  or  $N_c$ .

TABLE II  
 RESULTS OF SUBCRITICAL MEASUREMENTS WITH 1.8 wt % Pu-Al RODS IN LIGHT WATER  
 (Light water temperature: 17 C)

Lattice Spacing, Inches	H/Pu, Atom Ratio	H <sub>2</sub> O/Rod, Volume Ratio	Approach-to-critical Measurements		Exponential Measurements		Extrapolation Length <sup>***</sup> (λ), cm	Buckling B <sub>c</sub> <sup>2</sup> , 10 <sup>-6</sup> cm <sup>-2</sup> ***
			Critical Number of 43.83-in. Rods, Cylindrical Geometry*	Critical Mass, g Pu*	Number of Rods	Slope (γ) of Vertical Traverse, cm <sup>-1</sup> **		
0.75	630	0.9361	563.4 ± 0.9	4011 ± 6.4	348	0.03996 ± 0.00014	8.89 ± 0.05	6024 ± 20
0.80	810	1.203	510.3 ± 0.9	3633 ± 6.4	312	0.04179 ± 0.00022	8.26 ± 0.07	6127 ± 28
0.85	1001	1.487	493.9 ± 1.0	3517 ± 7.1	312	0.03992 ± 0.00019	7.56 ± 0.06	6009 ± 26
0.90	1204	1.788	515.8 ± 1.0	3672 ± 7.1	300	0.04154 ± 0.00032	7.96 ± 0.12	5272 ± 32
0.95	1418	2.106	577.7 ± 1.2	4113 ± 8.5	366	0.03413 ± 0.00019	7.07 ± 0.06	4734 ± 23

\* Each error is the RMS deviation of measurements from four detectors.

\*\* The errors are a measure of the goodness of fit to a straight line from at least 10 points as determined by a least squares analysis.

\*\*\* The errors are from \* and \*\* combined.

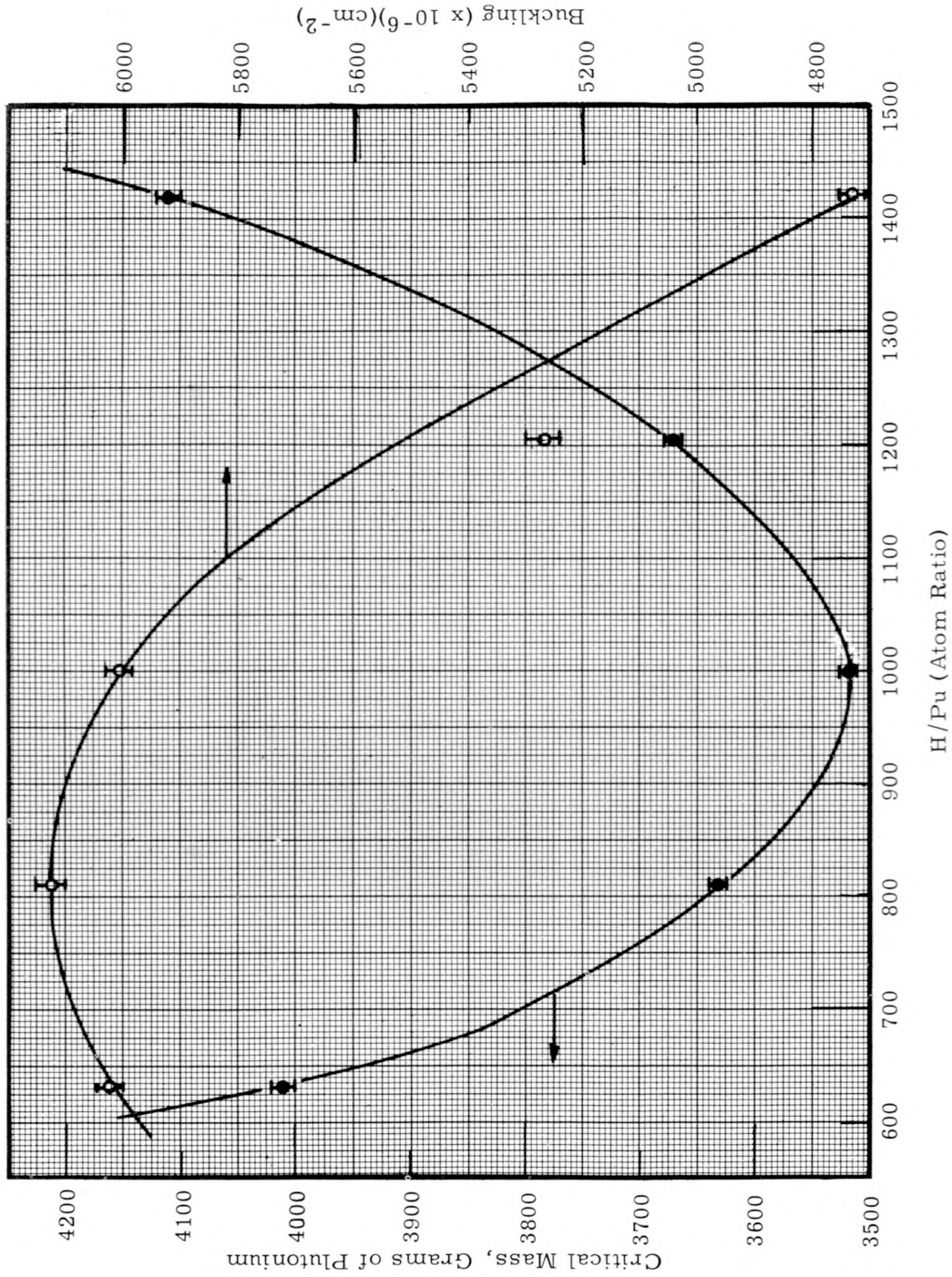


FIGURE 2

Critical Mass and Buckling Data for 1.8 wt % Pu-Al  
(Pu<sup>240</sup> content is 5.58 percent of total plutonium.)

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EXPERIMENTAL CRITICAL MASS PHYSICS

Critical Mass Experiments with Plutonium-Nitrate Solutions

R. C. Lloyd and E. D. Clayton

A series of criticality experiments was begun with plutonium-nitrate solutions in a 14-inch diameter stainless steel sphere, fully reflected with water. Plutonium concentrations were in the range of  $\sim 38$  to  $46$  g Pu/ $\ell$  with an acid molarity of  $\sim 4$ .

The data obtained from the water reflected experiments at low plutonium concentrations will provide a needed tie point for comparing the current experimental results with the early Hanford P-11 experiments which were conducted with dilute plutonium solutions.<sup>(1)</sup>

The data from the current experiments are summarized in Table I, which follows. The results of other measurements are given in previous quarterly reports.<sup>(2-4)</sup> Further data will be obtained for solutions with nitric acid molarities of two and six in order to evaluate the effect of nitrate on the criticality of the water reflected systems.

Data were obtained for estimating the correction to the measured critical volumes for the effect of the 0.044-inch stainless steel vessel wall. For these measurements additional layers of stainless steel were added to the vessel in the form of thin nesting shells in thicknesses of 0.036 and 0.072 inch. The stainless steel shell reduces the effectiveness of the water reflector slightly. In terms of volume, the correction is  $9.5 \pm 1.5$  ml/mil of stainless steel thickness, or the critical volume would be  $\sim 0.4$   $\ell$  smaller in the absence of the stainless steel.

A mock-up of the sphere neck was positioned against the side of the vessel in one of the experiments; its effect on criticality was within the uncertainty of the measured critical volumes.

TABLE I  
CRITICALITY STUDIES WITH PLUTONIUM SOLUTIONS IN 14-INCH DIAMETER STAINLESS STEEL SPHERE\*

Experiment Number	Date	Reflector	Plutonium, g/l	Hydrogen <sup>+</sup> , g/l	Specific Gravity	H <sub>2</sub> O, g/l	Total NO <sub>3</sub> , g/l	H/Pu Ratio	Critical Volume, Liters	Critical Mass, kg
1142090	6-5-62	0.044-in. SS + water	46.0	4.22	1.207	840	306	506.7	21.1 <sup>+ 0.02</sup> - 0.04	0.97
1142091	6-7-62	0.044-in. SS + water	42.1	4.22	1.203	850	302	559.9	22.0 <sup>+ 0.06</sup> - 0.08	0.93
1142094	6-12-62	0.044-in. SS + water	39.2	4.20	1.198	843	313	596.5	23.1 <sup>+ 0.05</sup> - 0.06	0.91
1142095	6-14-62	0.08-in. SS + water	38.8	4.14	1.194	845	311	603.6	23.7 <sup>+ 0.12</sup> - 0.19	0.92
1142096	7-5-62	0.08-in. SS + water	39.9	4.12	1.199	857	297	594.9	23.1 <sup>+ 0.04</sup> - 0.05	0.92
1142097	7-9-62	0.116-in. SS + water	40.3	4.13	1.199	857	298	589.0	23.3 <sup>+ 0.07</sup> - 0.09	0.94
1142098	7-10-62	0.044-in. SS + water	40.4	4.07	1.199	856	295	586.5	22.6 <sup>+ 0.04</sup> - 0.05	0.91
1142099	7-11-62	0.044-in. SS + water + mock-up of sphere neck	40.4	4.07	1.199	856	295	586.5	22.6 <sup>+ 0.03</sup> - 0.03	0.91
1142100	7-12-62	0.044-in. SS + water	38.4	4.06	1.196	854	292	615.6	23.3 <sup>+ 0.04</sup> - 0.05	0.89

\* Measured Sphere Volume - 23.22 liters  
 Wall Thickness - 0.044 inch

From these measurements the vessel would be just critical when full (23.22  $\ell$ ) for a plutonium concentration of 38.6 g Pu/ $\ell$  with 292 g  $\text{NO}_3/\ell$ . The critical mass would be 0.90 kg plutonium.

In order to compare these data with the previous P-11 results, it is necessary to apply corrections for any differences in the isotopic content of the plutonium, and to make the comparisons at the appropriate plutonium and nitrate concentrations.<sup>(1)</sup> In the P-11 experiments some data were obtained with a 14-inch sphere in which the  $\text{Pu}^{240}$  content was 4.4 percent. The  $\text{Pu}^{240}$  content of the plutonium in the current criticality studies is 4.6 percent, and therefore, only a small correction is required because of the  $\text{Pu}^{240}$  effect.

The results of the measurements completed to date with the water reflected critical assembly indicate agreement with the early P-11 experiments. The critical mass values also appear to correspond roughly to those in which the 4-inch concrete reflector was used; a concrete reflector of 4-inch thus being equivalent to an effectively infinite water reflector.<sup>(4)</sup>

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Limiting Critical Concentration of Pu<sup>239</sup> and U<sup>235</sup>  
in Aqueous Solutions by Monte Carlo Techniques

C. R. Richey

The limiting critical concentration, i. e., the concentration for which the infinite multiplication factor is unity, of a plutonium-water mixture was previously determined to be  $8.4 \pm 1$  g Pu<sup>239</sup>/ℓ from  $k_{\infty}$  measurements in the Hanford PCTR, <sup>(1)</sup> whereas the limiting critical concentration has been calculated to be 7.25 g Pu<sup>239</sup>/ℓ utilizing the 9-Zoom Multi-Group Diffusion Code. <sup>(2)</sup> In an effort to resolve this difference, a series of Monte Carlo calculations were made of  $k_{\infty}$  for Pu<sup>239</sup>-water mixtures with the Homogeneous Infinite System Monte Carlo (HISMC) code. The limiting critical concentration of 93.15 percent (weight) enriched UO<sub>2</sub>F<sub>2</sub>-water solution was also calculated with HISMC to check the ability of the code to reproduce a well-determined experimental value. The critical concentration of enriched UO<sub>2</sub>F<sub>2</sub>-water solution was experimentally determined to be  $12.97 \pm 0.05$  g U/ℓ ( $12.1$  g U<sup>235</sup>/ℓ) from PCTR experiments and critical experiments at ORNL. <sup>(1, 3)</sup>

The infinite multiplication factor was computed for plutonium concentrations of 7.5, 8.4 and 11.4 g/ℓ and uranium concentrations of 12.97 and 13.5 g/ℓ. A total of approximately 3000 neutron histories were traced for each concentration studied. The infinite multiplication factor was determined from the number of fission and absorption events by:

$$k_{\infty} = \frac{\nu^{25} \times U^{235} \text{ Fissions} + \nu^{28} \times U^{238} \text{ Fissions}}{\text{Total Absorptions}} \quad (\text{Enriched Uranium Systems})$$

and

$$k_{\infty} = \frac{\nu^{49} \times \text{Pu}^{239} \text{ Fissions}}{\text{Total Absorptions}} \quad (\text{Pu}^{239} \text{ Systems})$$

where

$$\nu^{25} = 2.43 \pm 0.02$$

$$\nu^{49} = 2.89 \pm 0.03$$

$$\nu^{28} = 2.85 \pm 0.04.$$

The effect of  $\nu^{25}$  and  $\nu^{49}$  being a function of neutron energy was found to be negligible in the well-thermalized solutions studied and has been ignored in the calculations. A summary of the calculations is given in the following table.

MONTE CARLO RESULTS FOR ENRICHED  $UO_2F_2$  AND  $Pu^{239}$  SOLUTIONS

Enriched  $UO_2F_2$ -Water Solutions

Run No.	12.97 g U/l			13.5 g U/l		
	25 Fissions	28 Fissions	Absorptions	25 Fissions	28 Fissions	Absorptions
1	394.22	0.07	973.42	407.14	0.00	989.31
2	401.11	0.00	994.10	403.25	0.00	974.17
3	413.18	0.00	1011.58	427.45	0.00	1017.38
Totals	1208.51	0.07	2979.10	1237.84	0.00	2980.86
	$k_\infty = 0.986 \pm 0.012$			$k_\infty = 1.009 \pm 0.013$		

$Pu^{239}$ -Water Solution

Run No.	7.5 g $Pu^{239}$ /l		8.4 g $Pu^{239}$ /l		11.4 g $Pu^{239}$ /l	
	Fissions	Absorptions	Fissions	Absorptions	Fissions	Absorptions
1	336.99	996.69	351.77	989.62	420.40	1021.15
2	332.26	980.00	383.31	1040.84	402.86	996.52
3	362.81	1041.02	350.40	987.55	408.08	990.49
Totals	1032.06	3017.71	1085.48	3018.01	1231.34	3008.16
	$k_\infty = 0.988 \pm 0.018$		$k_\infty = 1.039 \pm 0.019$		$k_\infty = 1.183 \pm 0.013$	

The limiting critical concentrations obtained from a simple interpolation of the above results are given in the following table along with the values obtained experimentally.

LIMITING CRITICAL CONCENTRATION OF Pu<sup>239</sup> AND U<sup>235</sup>

<u>Method</u>	<u>Pu<sup>239</sup> (g/Pu<sup>239</sup>/ℓ)</u>	<u>U<sup>236</sup> (g U/ℓ)</u>
Monte Carlo	7.71 ± 0.46	13.29 ± 0.41
Experimental	8.4 ± 1.0	12.97 ± 0.05

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