MLM-1395

MASTER

NEUTRON MULTIPLICATION EXPERIMENT WITH PLUTONIUM-238 DIOXIDE DOUBLE SEALED IN CALORIMETER PRESSURE CONTAINERS

R. A. Wolfe, D. A. Edling, D. F. Giessing, J. B. Kahle, and W. F. Stubbins

AEC Research and Development REPORT

MONSANTO RESEARCH CORPORATION

A SUBSIDIARY OF MONSANTO COMPANY

MOUND LABORATORY

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.65

LEGAL NOTICE

MLM-1395
TID-4500
UC-46
Criticality Studies

NEUTRON MULTIPLICATION EXPERIMENT WITH PLUTONIUM-238 DIOXIDE DOUBLE SEALED IN CALORIMETER PRESSURE CONTAINERS

R. A. Wolfe
D. A. Edling
D. F. Giessing*
J. B. Kahle
W. F. Stubbins**

Date: January 9, 1967
Issued: May 15, 1967

*U.S. Atomic Energy Commission
Operational Safety Division
Albuquerque Operations Office
Albuquerque, New Mexico

**Department of Physics
University of Cincinnati
Cincinnati, Ohio

MONSANTO RESEARCH CORPORATION
A Subsidiary of Monsanto Company

MOUND LABORATORY
Miamisburg, Ohio operated for

UNITED STATES ATOMIC ENERGY COMMISSION

U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

ABSTRACT

The neutron multiplication of high isotopic analysis plutonium dioxide assembled in various planar arrays in both water and air media was determined. The isotopic distribution of the plutonium was approximately 80% ^{238}Pu, 16% ^{239}Pu, 3% ^{240}Pu, and 1% ^{241}Pu. Twenty-one double-sealed calorimeter pressure containers, each containing approximately 130 grams of the plutonium-238 isotope, were assembled in three planar arrays consisting of 3-in., 1-in., and 1/16-in. edge-to-edge spacing. No significant neutron multiplication was detectable in the fully assembled 1-in. planar array in the water medium or in the 3-in. planar array in the air medium. A low multiplying region ($M \approx 1.025$) did exist in the 1/16-in. planar array but was not large enough to be used in reliably extrapolating to the critical mass of plutonium-238.

INTRODUCTION

To determine the nuclear safety of the plutonium-238 isotope, a neutron multiplication experiment was performed with plutonium dioxide double sealed in calorimeter pressure containers. The isotopic distribution of the plutonium was approximately 80% ^{238}Pu, 16% ^{239}Pu, 3% ^{240}Pu, and 1% ^{241}Pu. Twenty-one pressure containers, each containing approximately 130 grams of plutonium-238 isotope, were assembled in three planar arrays consisting of 3-in., 1-in., and 1/16-in. edge-to-edge spacing. The experiment was performed in both air and water media.

EXPERIMENTAL RESULTS IN A WATER MEDIUM

The experiments were performed in water to determine whether the neutron count rate would increase due to multiplication as the containers were brought to closer edge-to-edge spacings, thus changing the water medium properties from a moderator to a reflector. The two edge-to-edge spacings evaluated in this experiment were 1-in. and 1/16 in., respectively. The experimental arrangement is shown in Figure 1.

The recorded neutron count rate of each container was measured individually in its array location with BF_3 and ^3He proportioned detectors in their respective positions as shown in Figure 1. Empty calorimeter containers were used during the array increase to correct for the neutron scattering and reflection properties of the loaded containers. The loaded containers were added one at a time, each to its unique preassigned location, to build up the array. The neutron yield was measured with each increment of added containers and a neutron multiplication calculated as follows:

$$M = \frac{\text{Recorded Count Rate - Background}}{\text{Accumulative Count Rate - Background}}$$

The experimental data and results for the 1-in. and the 1/16-in. arrays are tabulated in Tables 1 and 2, respectively. A plot of the neutron multiplication obtained for each array is shown in Figures 2 and 3. The multiplication obtained with the two detectors was averaged and the statistical error assigned to the average value. The results shown in Figure 2 for the 1-in. array indicated that no significant neutron multiplication existed. The multiplication shown in Figure 3 for the 1/16-in. spaced array tends to be slightly higher ($M \approx 1.02$) than observed in the 1-in. spaced array.

It was anticipated that a moderated system might show a depression of the neutron multiplication due the large capture cross section of the ^{238}Pu isotope for thermal neutrons. As shown in Figure 2, no depression in the neutron yield was observed. The results reported in Figure 3 did not confirm whether the water performed as a significant neutron reflector.

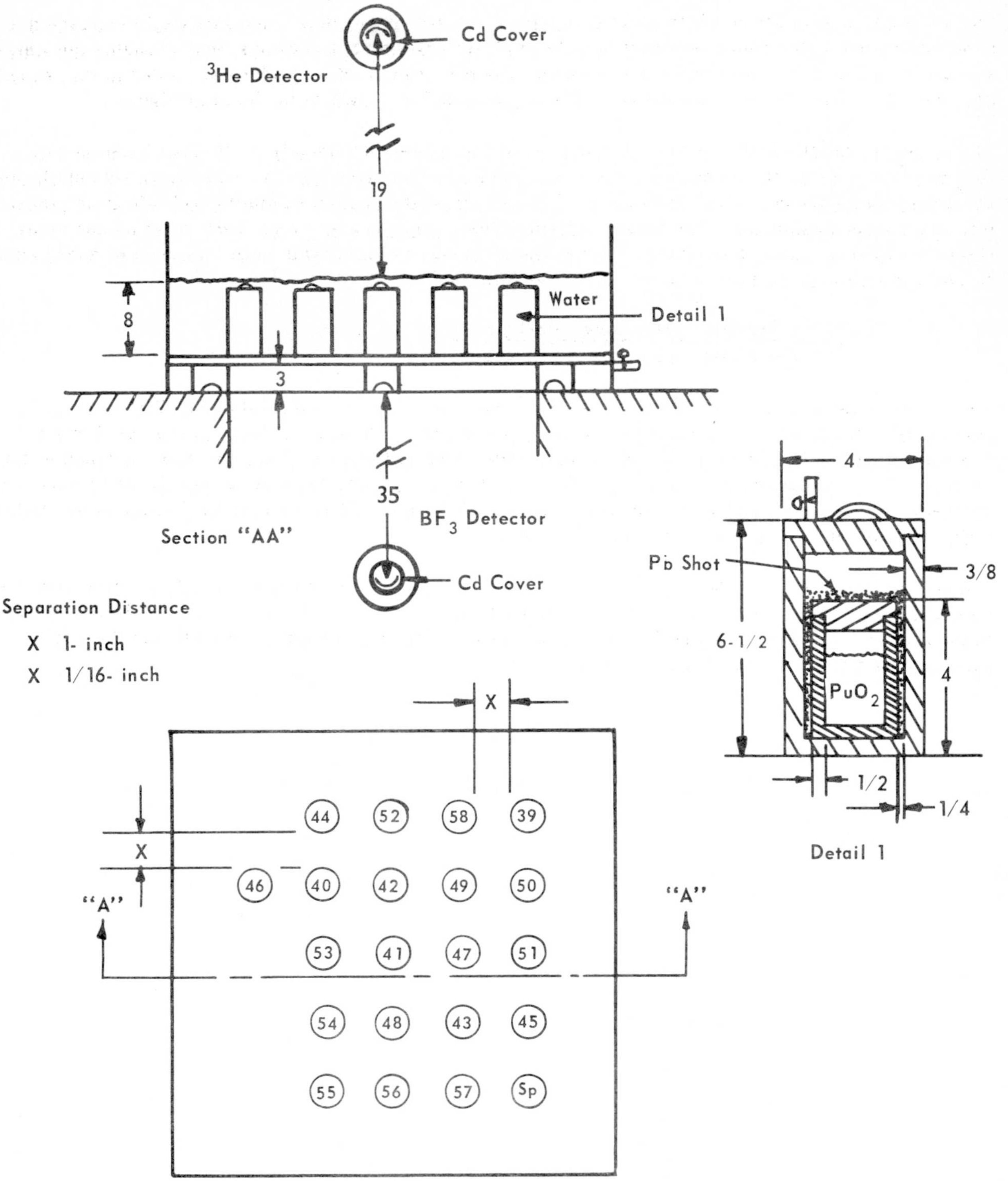

Figure 1. *Neutron Multiplication Experimental Arrangement for ^{238}Pu O_2 Storage Capsules in a Water Medium. (Dimensions in Inches)*

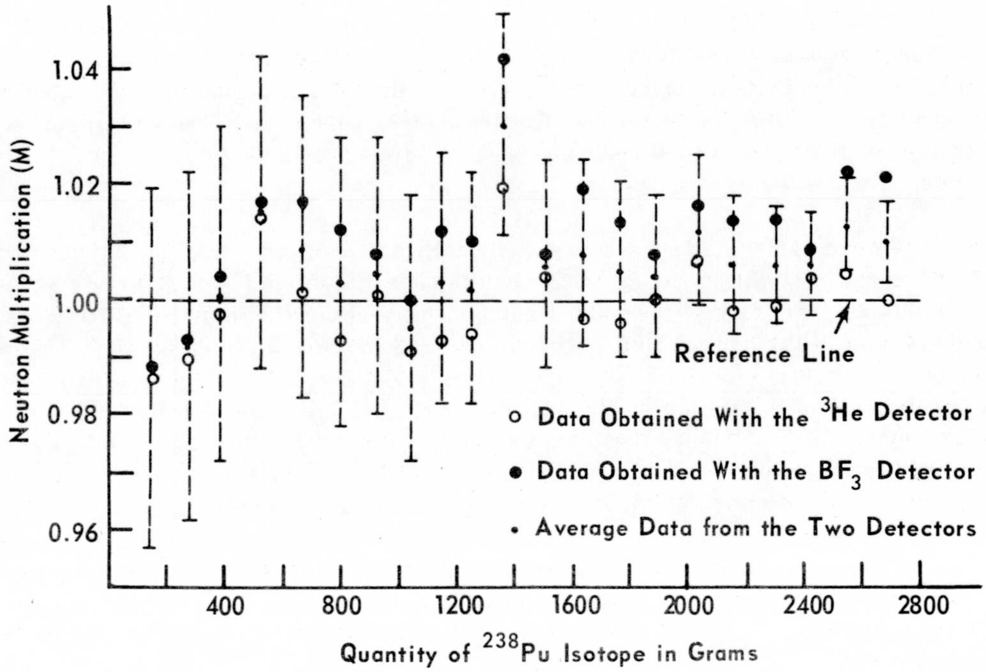

Figure 2. *Neutron Multiplication of Calorimeter Cans Containing $^{238}Pu\ O_2$ as a Function of the Quantity of ^{238}Pu Isotope When Assembled in a Planar Array of One Inch Edge-to-Edge Spacing in a Water Medium.*

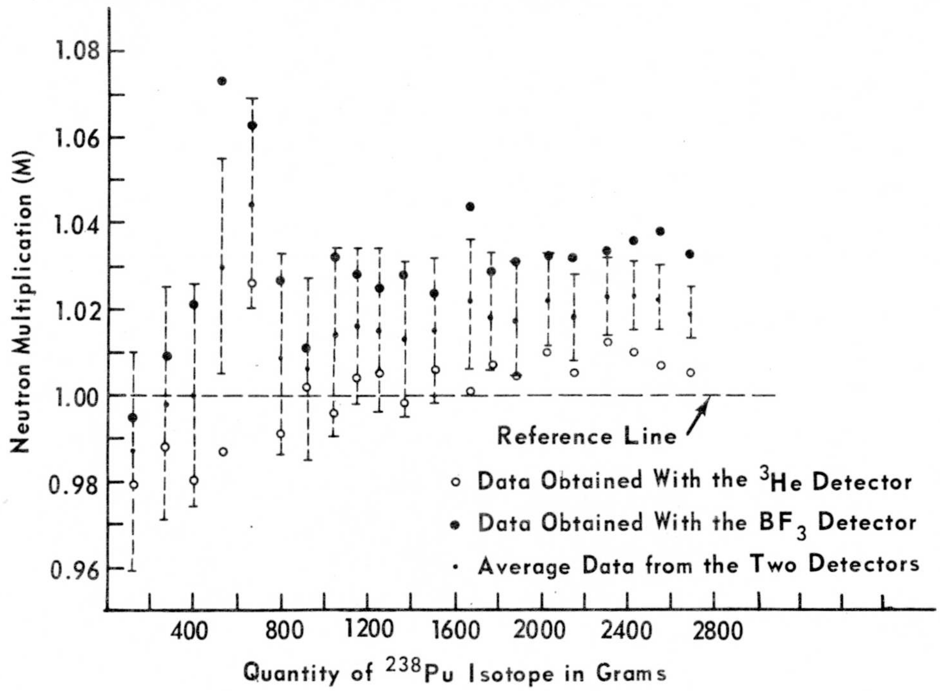

Figure 3. *Neutron Multiplication of Calorimeter Cans Containing $^{238}Pu\ O_2$ as a Function of the Quantity of ^{238}Pu Isotope When Assembled in a Planar Array of 1/16-Inch Edge-to-Edge Spacing in a Water Medium.*

EXPERIMENTAL RESULTS IN AN AIR MEDIUM

A fast system is the most probable critical system that exists for the plutonium-238 isotope. A thermal system is not probable because of the large thermal neutron capture cross section and the small thermal fission cross section. The experiment performed in air was to determine the neutron multiplication as a function of the quantity of ^{238}Pu isotope for various planar array spacings. It was anticipated that the results would tend to indicate the critical mass of ^{238}Pu and thus provide a guide to the safe storage, processing, and capsule loading of this isotope.

The two edge-to-edge spacings evaluated in this experiment were 3-in. and 1/16-in., respectively. The experimental arrangement is shown in Figure 4. The neutron multiplication for each array was determined by the same technique as described for the water medium. The experimental data and results are tabulated in Tables 3 and 4. A plot of the neutron multiplication obtained for each array is shown in Figures 5 and 6. The results shown in Figure 5 for the 3-in. spacing array indicated that no neutron multiplication was observed. The results shown in Figure 6 for the 1/16-in. array indicated that a small neutron multiplication existed of approximately 1.025. Although these data exist in a low multiplying region, they do indicate that the increase in neutron count rate was due to fissioning of the ^{238}Pu isotope. These results illustrate the increase in neutron multiplication as the more favorable geometry was obtained.

This was the first attempt ever to assemble this quantity of ^{238}Pu into a systematic array under controlled conditions. These results are significant in determining the nuclear safety of ^{238}Pu and will be used as a guide by the Mound Laboratory Criticality Committee in establishing and approving storage array designs, operating procedures, etc.

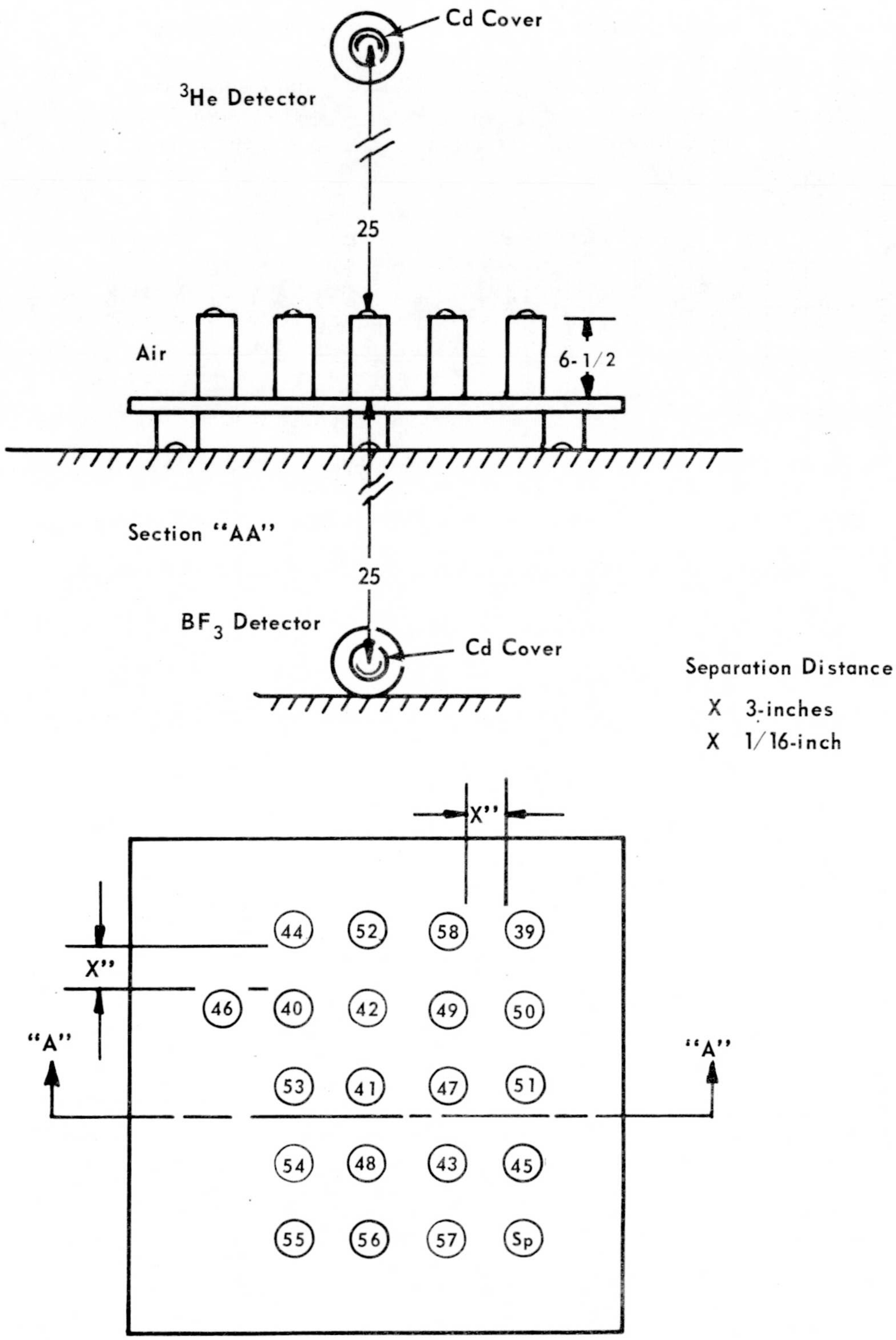

Figure 4. *Neutron Multiplication Experimental Arrangement for $^{238}PuO_2$ Storage Capsules in an Air Medium. (Dimensions in Inches)*

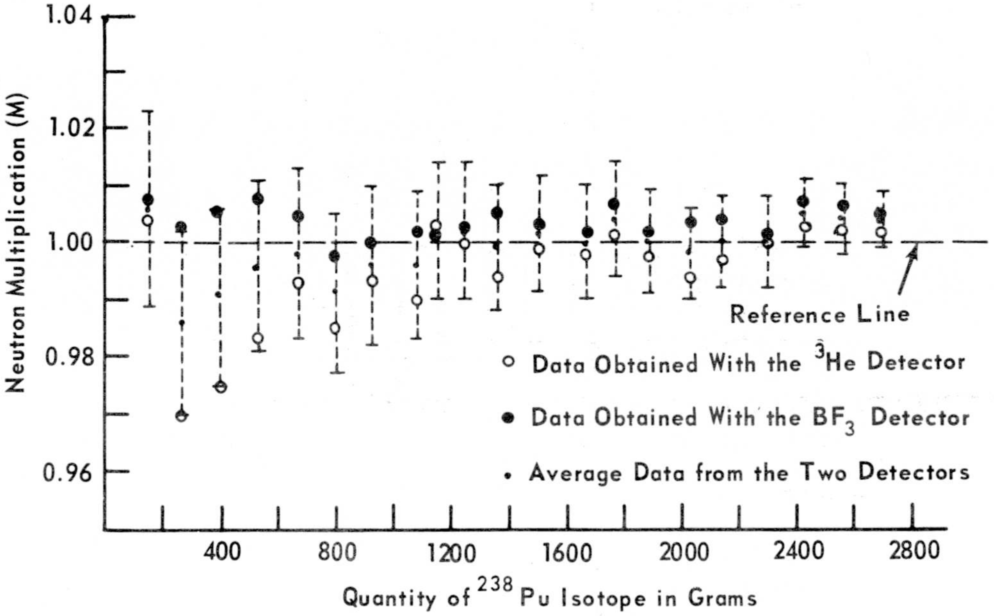

Figure 5. *Neutron Multiplication of Calorimeter Cans Containing ^{238}Pu O_2 as a Function of the Quantity of ^{238}Pu Isotope When Assembled in a Planar Array of 3-inch Edge-to-Edge Spacing in an Air Medium.*

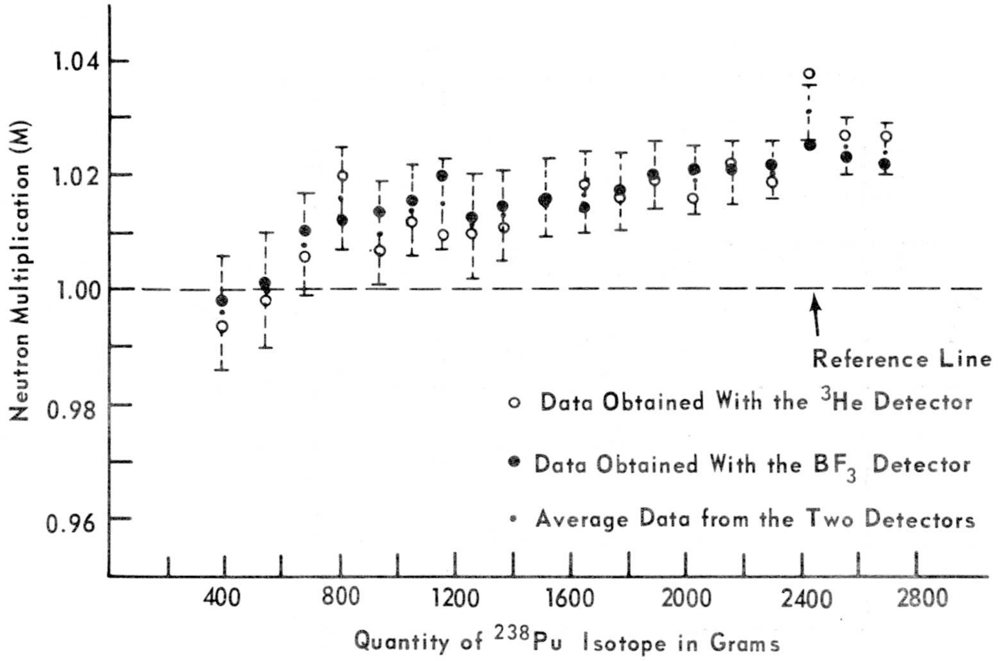

Figure 6. *Neutron Multiplication of Calorimeter Cans Containing ^{238}Pu O_2 as a Function of the Quantity of ^{238}Pu Isotope When Assembled in a Planar Array of 1/16-inch Edge-to-Edge Spacing in an Air Medium.*

CONCLUSIONS

Based upon the results obtained in these experiments, the following conclusions are drawn:

(1) No significant neutron multiplication was detectable in the fully assembled 1-in. planar array in the water medium or in the 3-in. planar array in the air medium.

(2) A small neutron multiplication ($M \approx 1.025$) existed in the fully assembled 1/16-in. planar array in both the water and the air media.

(3) The results showed a low multiplying region did exist in the 1/16-in. planar arrays but was not large enough to be used in reliably extrapolating to the critical mass of plutonium-238.

(4) The results confirmed that no criticality hazard exists with approximately 2.7 kilograms of ^{238}Pu when assembled in the fuel configurations and planar arrays evaluated. This does not mean that 2.7 kilograms of ^{238}Pu would not be hazardous if assembled in a more favorable geometry, such as a solid sphere.

Table I

NEUTRON MULTIPLICATION EXPERIMENTAL DATA FOR THE I-INCH SPACING ARRAY IN A WATER MEDIUM

	Detector 1 ^3He				Detector 2 BF_3			
No. of Containers	Accum. Quantity of Isotope Σ(g)	Accum. Count Rate Σ(C-B) (cts/min)	Recorded Count Rate (C-B) (cts/min)	Multiplication M	Accum. Count Rate Σ(C-B) (cts/min)	Recorded Count Rate (C-B) (cts/min)	Multiplication M	Average Multiplication From Two Detectors ($\bar{M}$)
41	146	3770	3722	0.987	4429	4377	0.988	0.988±.031
41 + 47	274	10038	9968	0.993	10411	10304	0.990	0.992±.030
49	398	14065	14127	1.004	15288	15251	0.998	1.001±.029
42	537	19335	19609	1.014	20940	21286	1.017	1.015±.027
48	667	24674	24706	1.001	26220	26669	1.017	1.009±.026
43	800	29579	29369	0.993	30235	30603	1.012	1.003±.025
45	927	32518	32558	1.001	34074	34333	1.008	1.004±.024
51	1044	35965	35625	0.991	38294	38309	1.000	0.995±.023
50	1155	38648	38377	0.993	42386	42911	1.012	1.003±.021
39	1254	40862	40630	0.994	47014	47462	1.010	1.002±.020
58	1362	43387	45207	1.042	51428	52420	1.019	1.030±.019
52	1502	46624	46826	1.004	56135	56569	1.008	1.006±.018
44	1637	49945	49749	0.996	60641	61783	1.019	1.008±.016
40	1769	54238	54000	0.996	65617	66500	1.013	1.005±.015
53	1897	58560	58553	1.000	70366	70914	1.008	1.004±.014
54	2033	62906	63321	1.007	74535	75768	1.017	1.012±.013
55	2158	66286	66168	0.998	78485	79603	1.014	1.006±.012
56	2301	69874	69796	0.999	82883	84083	1.014	1.006±.010
57	2424	72950	73261	1.004	86632	87368	1.009	1.006±.009
Sp	2556	75372	75704	1.004	89955	91924	1.022	1.013±.008
46	2691	78125	78159	1.000	93977	95978	1.021	1.010±.007

Table 2

NEUTRON MULTIPLICATION EXPERIMENTAL DATA FOR THE 1/16-INCH SPACING ARRAY IN A WATER MEDIUM

		Detector 1 ^3He			Detector 2 BF_3			
No. of Containers	Accum. Quantity of Isotope Σ(g)	Accum. Count Rate Σ(C-B) (cts/min)	Recorded Count Rate (C-B) (cts/min)	Multiplication (M)	Accum. Count Rate Σ(C-B) (cts/min)	Recorded Count Rate (C-B) (cts/min)	Multiplication (M)	Average Multiplication From Two Detectors ($\bar{M}$)
47	128	5340	5227	0.979	4963	4938	0.995	0.987±.028
47 + 41	274	13395	13239	0.988	11256	11355	1.009	0.998±.027
49	398	18761	18385	0.980	16486	16828	1.021	1.000±.026
42	537	25670	25325	0.987	22710	24364	1.073	1.030±.025
48	667	32987	35072	1.063	28528	29285	1.026	1.044±.024
43	800	39620	39271	0.991	33174	34066	1.027	1.009±.023
45	927	43954	44059	1.002	37558	37953	1.011	1.006±.021
51	1044	48646	48455	0.996	42058	43414	1.032	1.014±.020
50	1155	52467	52711	1.004	46663	47969	1.028	1.016±.019
39	1254	55793	56090	1.005	51778	53074	1.025	1.015±.018
58	1362	59308	59181	0.998	56581	58181	1.028	1.013±.017
52	1502	63876	64273	1.006	61593	63081	1.024	1.015±.016
44	1637	68326	68420	1.001	66745	69718	1.044	1.022±.015
40	1769	74024	74647	1.008	72095	74201	1.029	1.018±.014
53	1897	79788	80131	1.004	77073	79448	1.031	1.017±.012
54	2033	85568	86443	1.010	82030	84710	1.033	1.022±.011
55	2158	90275	90762	1.005	86493	89288	1.032	1.018±.010
56	2301	95615	96810	1.012	91417	94539	1.034	1.023±.009
57	2424	100248	101176	1.010	95604	99039	1.036	1.023±.008
Sp	2556	104061	104803	1.007	99649	103526	1.038	1.022±.007
46	2691	107877	108355	1.005	104230	107626	1.033	1.019±.006

Table 3

NEUTRON MULTIPLICATION EXPERIMENTAL DATA FOR THE 3-INCH SPACING ARRAY IN AN AIR MEDIUM

		Detector 1 ^3He			Detector 2 BF_3			
No. of Containers	Accum. Quantity of Isotope Σ(g)	Accum. Count Rate Σ(C-B) (cts/min)	Recorded[a] Count Rate (C-B) (cts/min)	Multiplication M	Accum. Count Rate Σ(C-B) (cts/min)	Recorded[b] Count Rate (C-B) (cts/min)	Multiplication M	Average Multiplication From Two Detectors ($\bar{M}$)
41	146	9849	9889	1.004	22286	22475	1.008	1.006±.011
41 + 47	274	17891	17354	0.970	39913	40029	1.003	0.986±.016
49	398	25586	24950	0.975	58808	59152	1.006	0.991±.016
42	537	34733	34164	0.984	81058	81756	1.008	0.996±.015
48	667	44229	43898	0.993	100844	101330	1.005	0.998±.015
43	800	53472	52692	0.985	117496	117269	0.998	0.991±.014
45	927	61730	61289	0.993	132241	132342	1.000	0.996±.014
51	1044	69407	68730	0.990	148220	148549	1.002	0.996±.013
50	1155	75790	75954	1.003	162540	162934	1.002	1.002±.012
39	1254	83882	83922	1.000	180333	180818	1.003	1.002±.012
58	1362	90527	89955	0.994	197315	198252	1.005	0.999±.011
52	1502	97890	97745	0.999	215587	216231	1.003	1.002±.010
44	1637	105247	104916	0.997	233466	233851	1.002	0.999±.010
40	1769	112675	112756	1.001	250859	252535	1.007	1.004±.009
53	1897	119868	119526	0.997	266515	267138	1.002	1.000±.009
54	2033	127606	126847	0.994	281070	281984	1.003	0.998±.008
55	2158	135471	135044	0.997	293790	294977	1.004	1.000±.008
56	2301	143885	143920	1.000	307492	307905	1.001	1.000±.007
57	2424	151542	151930	1.003	320086	322183	1.007	1.005±.006
Sp	2556	159160	159594	1.003	331890	333909	1.006	1.004±.006
46	2691	165190	165523	1.002	345476	347216	1.005	1.004±.005

NOTE: [a]The dead time correction was not significant for the ^3He detector.

[b]The dead time for the BF_3 detector was significant ($\tau = 6.88 \times 10^{-8}$ minutes). The appropriate correction has been applied to the recorded count rate.

Table 4

NEUTRON MULTIPLICATION EXPERIMENTAL DATA FOR THE 1/16-INCH SPACING ARRAY IN AN AIR MEDIUM

	Detector 1 ^3He				Detector 2 BF_3			
No. of Containers	Accum. Quantity of Isotope Σ(g)	Accum. Count Rate Σ(C-B) (cts/min)	Recorded[a] Count Rate (C-B) (cts/min)	Multiplication M	Accum. Count Rate Σ(C-B) (cts/min)	Recorded[b] Count Rate (C-B) (cts/min)	Multiplication M	Average Multiplication From Two Detectors ($\bar{M}$)
47	128	9264	- -	- -	20840	- -	- -	- -
47 + 41	274	21437	- -	- -	47405	- -	- -	- -
49	398	30947	30758	0.994	69629	69498	0.998	0.996±.010
42	537	42267	42179	0.998	95211	95342	1.001	1.000±.010
48	667	54152	54473	1.006	119900	121120	1.010	1.008±.009
43	800	65742	67050	1.020	140404	142044	1.012	1.016±.009
45	927	74675	75165	1.007	159968	162127	1.014	1.010±.009
51	1044	83830	84863	1.012	180521	183496	1.016	1.014±.008
50	1155	91263	92186	1.010	198278	202147	1.020	1.015±.008
39	1254	99729	100703	1.010	221211	223979	1.013	1.012±.008
58	1362	107214	108414	1.011	240630	244221	1.015	1.013±.008
52	1502	115463	117349	1.016	261519	265645	1.016	1.016±.007
44	1637	123835	126080	1.018	283293	287469	1.015	1.017±.007
40	1769	132593	134735	1.016	304281	309551	1.017	1.017±.007
53	1897	141132	143763	1.019	323991	330505	1.020	1.020±.006
54	2033	149889	152262	1.016	343192	350274	1.021	1.019±.006
55	2158	158051	161606	1.022	361326	369035	1.021	1.022±.006
56	2301	167248	170369	1.019	380496	388947	1.022	1.021±.005
57	2424	175832	182510	1.038	398196	408218	1.025	1.031±.005
Sp	2556	184022	189034	1.027	415637	425309	1.023	1.025±.005
46	2691	191070	196302	1.027	433403	442900	1.022	1.024±.004

NOTE: [a]The dead time correction was not significant for the ^3He detector.

[b]The dead time for the BF_3 detector was significant ($\tau = 6.88\times10^{-8}$ minutes). The appropriate correction has been applied to the recorded count rate.