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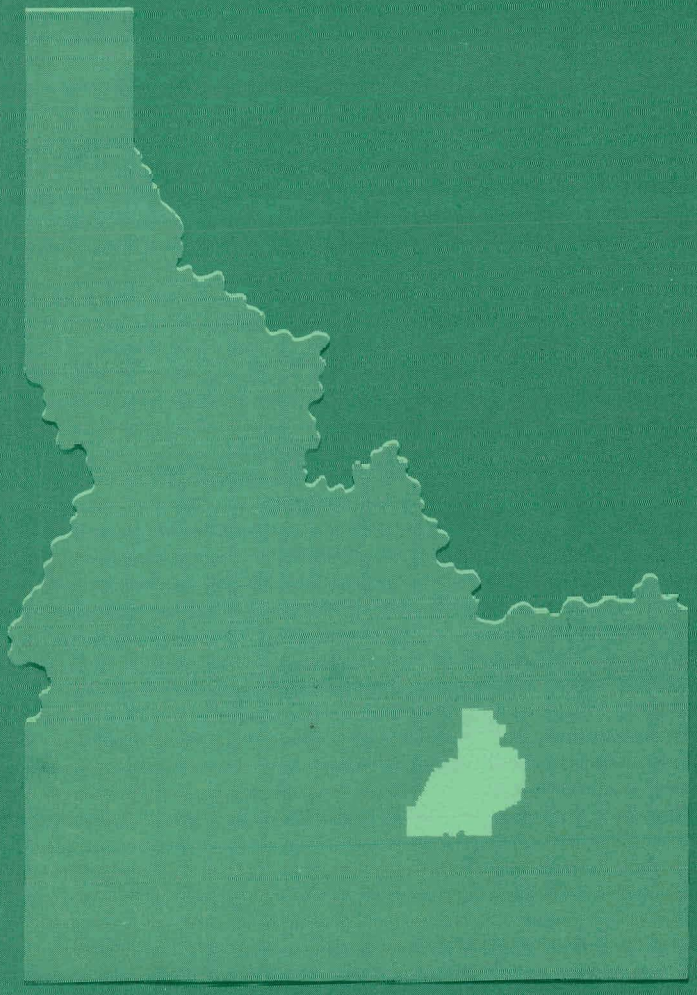
ADDITIONAL MEASUREMENTS OF THE REACTIVITY TRANSIENT IN IRRADIATED THORIUM

R. G. Nisle
D. A. Millsap

March 13, 1961



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SUMMARY

In a previous report the decay of Pa-233 into U-233 was traced by a series of measurements of reactivity in the Reactivity Measurement Facility (RMF).

The work reported herein consists of a similar experiment on two pair of thorium slugs. One pair of Savannah River type slugs were irradiated in the MTR in L-58 for an estimated 2.5 x 10^{20}nvt at an estimated flux level of 1.8 x 10^{14}nv. The other pair were Hanford type and were irradiated in A-5 an estimated 6.6 x 10^{20}nvt at an estimated level of 2.0 x 10^{13}nv.

Measurements in RMF were made in two positions. Estimates of the decay constant of Pa-233 and the U-233 content of the slugs are made. From the results it is concluded that (1) the long term reactivity transient is due to Pa-233 decay, (2) thorium slugs can be irradiated in the MTR at flux levels of 10^{14}nv to a U-233 content of 6 grams without damage to the slugs, and (3) the in-pile cross section of Pa-233 can be measured by this method provided an independent measurement of the irradiating flux is made.

CONTENTS

LIST OF TABLES

LIST OF FIGURES

ADDITIONAL MEASUREMENTS OF THE REACTIVITY TRANSIENT IN IRRADIATED THORIUM

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I. INTRODUCTION

In a previous report[1] the decay of Pa-233 into U-233 was traced by a series of measurements of reactivity in the Reactivity Measurement Facility (RMF). The work reported herein consists of a similar experiment.

Reactivity measurements were made on one pair of Hanford type thorium slugs during the period January, 1959 to December, 1959. They had been irradiated in position A-5 in the MTR for an estimated 6.6×10^{20} nvt.

The second pair were Savannah River type. Reactivity measurements were made on them during the period of August, 1959 to April, 1960. They had been irradiated in position L-58 in the MTR for an estimated 2.5×10^{20} nvt. There was no evidence of any damage to them as a result of irradiation in a high flux position such as L-58.

The results of this experiment yield a value for the decay constant of Pa-233 and a rough estimate of the irradiation flux. Further support is also given to the assumption, made in the previous report, that the fission products of U-233 contained in the slug have a negligible effect on the reactivity transient. This experiment gives no information about the total poison contribution of such fission products.

II. DESCRIPTION OF THE EXPERIMENT

The two Savannah River type slugs had previously received a considerable amount of irradiation in the MTR. They had been removed from the MTR in May, 1959 and were being stored in the canal to await the Pa-233 decay before measurement in the RMF. Hence, about 65 days elapsed between previous irradiation and the insertion into L-58. The Pa-233 from the previous irradiation was, therefore, nearly all decayed out.

The two Hanford type slugs were fresh slugs when they were inserted in the MTR, and the entire irradiation was accomplished in position A-5. The locations in the MTR during irradiation are shown in Figure 1.

Reactivity measurements were begun four days after removal from the MTR in the case of the Savannah River type slugs and two days after removal in the case of the Hanford type slugs. Thus, reactivity changes due to xenon-135 and samarium-149 were reduced.

During the measurement period the RMF was dismantled, reassembled, and recalibrated, all of which was necessitated by certain canal repairs. Consequently, the raw data required an adjustment to account for two different regulating rod calibrations. This was accomplished through the intermediary of standard slugs, one of which was unirradiated and one of which had previously been irradiated and allowed to cool for about four years.

Measurements were made in positions 1 and 6 as indicated in Figure 2. Because of the high level of U-233 that had been built up in these slugs the reactivity effect was larger than could be accommodated by the RMF regulating rod. Therefore, in order to make a measurement on these slugs it was necessary to use an intermediate standard containing some U-233. An irradiated slug that had been out of the reactor for several years was used for this purpose. Each set of measurements, therefore, consisted of measurements on the reactivity of the unirradiated and intermediate standards (net to the empty reactor) and on the reactivity of the subject slugs, net to the intermediate standard. Hence, the total reactivity effect due to the U-233 content was the sum of the effects.

III. TREATMENT OF THE DATA

Each day's data consisted of at least three readings on all slugs. These readings were averaged and the results are given in Table I. It will be noted that there is a considerable variation in the reactivity worth of the standard, even though there is no reason to believe that anything has happened to it to cause such variations. Hence, such variations are attributed to shifts in reactor calibration, and the raw data were corrected by means of the following formula

$$R_c = \frac{R_i}{S_i} \; S_c \qquad (1)$$

where R_c is the corrected reactivity worth of a slug.
S_c is an average of the reactivity worth of the intermediate standard.
R_i is average reactivity worth of a slug for readings taken on the i-th day. (Table I)
S_i is the average reactivity worth of the intermediate standard for readings taken on the i-th day. (Table I)
R_c, S_c, R_i, and S_i are all taken as net to the unirradiated slug.

Table II lists the reactivity worths in μk after being corrected in this manner. Figures 3 and 4 are graphs drawn from the data in Table II. In order to derive a value of the Pa-233 decay constant

from these data, a least squares fit of the following equation was made:

$$R(t) - R(o) = \left[R(\infty) - R(o)\right] \cdot \left[1 - \exp(-\lambda t)\right] \qquad (2)$$

In order to use this equation it was necessary to estimate a value for the reactivity at time zero, R(o). This was done by graphical extrapolation. The IBM 650 was programmed to find $R(\infty)$ and λ by an iterative process when t and R(t) are given. It was found that the process was convergent and about one-half hour machine time was required for a solution.

IV. RESULTS

Table III lists the results of these calculations. The half life of Pa-233 as derived from these data is seen to be 26.5 and 25.5 days. This compares favorably with the literature value of 27.4 days. The standard deviations listed in the last column of Table III refer to the deviations of the values listed in Table II from those calculated by the IBM 650. The data taken in position 1 are consistently better than those taken in position 6. This is attributed to the fact that the flux gradients in this position are much greater than they are in position 1 and hence any positioning error is more conducive to a reactivity error.

The U-233 content of these slugs was estimated at t = 0 and at the end of the decay period by means of the correlation given in IDO-16402 [2] (Figure 4 of that report is reproduced here as Figure 5). The results are given in Table IV. If an independent measurement of the flux were available it would be possible to calculate an in-pile cross section of Pa-233 from these data. By an IBM calculation similar to that described in IDO-16504 [3], a rough estimate of the flux in the two cases was made from the data in Table IV. The flux level for the Savannah River type slugs in the L-58 position was estimated to be about 1.8×10^{14}nv and for the Hanford type slugs in A-5 it was estimated to be about 2×10^{13}nv.

V. CONCLUSIONS

The results reported herein lead to the following conclusions:

(1) The long term reactivity transient observed in irradiated thorium is due to the decay of Pa-233 and the presence of U-233 fission products produces a negligible effect, since the measured half life was nearly the accepted value for Pa-233.

(2) Savannah River type thorium slugs can be irradiated in the MTR at flux levels of 10^{14}nv until the U-233 content has been built up to 6 grams without damage to the slugs.

(3) The in-pile cross section of Pa-233 can be measured by this method provided an independent measurement of irradiating flux is made.

VI. ACKNOWLEDGMENTS

The authors wish to express their appreciation to the RMF Staff for their assistance in making measurements on these slugs.

VII. REFERENCES

(1) Nisle, R. G., An Analysis of a Reactivity Transient in Irradiated Thorium. IDO-16513 (July 9, 1959)

(2) Nisle, R. G. and Fast, E., Progress Report on the Study of Irradiated Thorium. IDO-16402 (August 28, 1957)

(3) Nisle, R. G., Calculation of Isotope Production by the Cyclic Irradiation of Thorium. IDO-16504 (March 26, 1959)

TABLE I

AVERAGED REACTIVITY DATA BY DAYS

Savannah River Type Slugs

$\Delta \rho$ in μk
(Net to Unirradiated Slug)

		R_1				S_i	
		Slug 009E6		Slug 590D1		Intermediate Standard	
Date	Days	Pos. 1	Pos. 6	Pos. 1	Pos. 6	Pos. 1	Pos. 6
8/13/59	4	1187.3	1341.4	1152.5	1306.0	850.8	961.7
8/17	8	1200.7	1352.4	1163.0	1310.0	849.5	968.0
8/21	12	1216.1	1368.9	1174.4	1324.7	849.7	957.8
8/25	16	1230.9	1386.0	1187.6	1339.9	851.6	960.0
8/28	19	1234.8	1396.6	1188.9	1347.0	848.9	959.9
9/4	26	1257.5	1407.9	1208.8	1352.7	852.7	951.1
9/11	33	1275.8	1437.1	1223.0	1378.5	854.0	960.6
9/25	47	1295.8	1460.5	1237.5	1395.6	851.8	958.6
10/16	68	1325.6	1486.4	1263.5	1416.4	857.6	960.9
11/6	89	1342.7	1490.3	1278.9	1429.3	861.7	959.0
12/2	115	1343.1	1520.8	1276.6	1445.5	855.2	967.3
2/24/60	199	1393.8	1579.8	1326.6	1504.1	880.2	1002.0
3/4	208	1387.9	1589.9	1318.6	1517.5	877.0	1007.9
4/12	247	1409.3	1592.8	1338.5	1517.1	890.3	1005.9

TABLE I. (Continued)

AVERAGED REACTIVITY DATA BY DAYS

Hanford Type Slugs

$\Delta \rho$ in μk
(Net to Unirradiated Slug)

		R_i				S_i	
		Slug No. 21-75		Slug No. 21-76		Intermediate Standard	
Date	Days	Pos. 1	Pos. 6	Pos. 1	Pos. 6	Pos. 1	Pos. 6
1/5/59	2	1197.5	1327.1	1115.9	1241.3	881.6	982.0
1/7	4	1206.8	1325.1	1128.1	1240.3	886.6	977.4
1/16	13	1215.7	1339.5	1136.0	1255.2	877.9	969.3
1/23	20	1233.9	1357.4	1155.0	1274.0	881.6	978.1
1/30	27	1248.0	1381.2	1162.5	1292.8	881.2	979.7
2/6	34	1236.9	1356.2	1155.3	1265.4	866.4	955.1
2/19	47	1252.3	1394.7	1172.4	1305.0	871.5	972.4
3/6	62	1253.8	1402.8	1173.5	1311.3	864.8	967.1
3/27	83	1261.1	1413.5	1179.7	1322.9	864.4	967.6
4/21	108	1253.4	1424.2	1172.1	1333.6	854.8	974.5
5/22	149	1265.8	1424.4	1184.8	1332.1	861.6	967.2
6/24	182	1277.6	1440.6	1192.8	1342.5	866.5	979.9
7/29	217	1263.6	1416.4	1178.5	1328.2	855.4	965.1
12/4	345	1283.1	1435.6	1200.3	1344.5	872.2	976.2

TABLE II.

CORRECTED REACTIVITY DATA

Savannah River Type Slugs

$\Delta \rho$ in μk

S_c (Position 1) = 855

S_c (Position 6) = 960

	009E6		590D1	
Days	Pos. 1	Pos. 6	Pos. 1	Pos. 6
4	1193	1339	1158	1304
8	1208	1341	1170	1299
12	1224	1372	1181	1327
16	1236	1386	1192	1340
19	1244	1397	1197	1347
26	1261	1421	1212	1365
33	1277	1436	1224	1378
47	1301	1463	1242	1398
68	1322	1485	1260	1415
89	1332	1492	1269	1431
115	1343	1509	1276	1435
199	1354	1514	1289	1441
208	1353	1514	1285	1445
247	1353	1520	1285	1448

Note: The reactivity values of $\Delta\rho$, in the body of the table are calculated from those in Table I by means of equation (1).

TABLE II. (Continued)

CORRECTED REACTIVITY DATA

Hanford Type Slugs

$\Delta \rho$ in μk

S_c (Position 1) = 870.5

S_c (Position 6) = 972.3

	Slug 21-75		Slug 21-76	
Days	Pos. 1	Pos. 6	Pos. 1	Pos. 6
2	1182	1314	1102	1229
4	1185	1318	1107	1234
13	1206	1344	1126	1259
20	1219	1350	1140	1266
27	1233	1371	1148	1283
34	1243	1381	1160	1288
47	1250	1394	1171	1305
62	1262	1411	1181	1318
83	1270	1421	1188	1329
108	1276	1421	1194	1330
149	1279	1432	1197	1339
182	1283	1429	1199	1332
217	1286	1427	1200	1338
345	1281	1430	1198	1339

Note: The reactivity values $\Delta\rho$, in the body of the table are calculated from those in Table I by means of equation (1).

TABLE III

MEASURED VALUES OF Pa-233 DECAY CONSTANT AND HALF LIFE

		R(o) in μk	R(∞) in μk	λ(IBM)	$\tau_{\frac{1}{2}}$ (IBM)		
Slug No.	Position	Estim.	IBM	IBM	Days	Days	Std.Dev.of Data
009E6	1	1178	1176	1354	0.02526	27.4	1.16μk
009E6	6	1316	1311	1516	0.02827	24.5	4.51
590D1	1	1148	1145	1287	0.02431	28.5	1.32
590D1	6	1286	1280	1445	0.02680	25.8	4.57
		Averages			0.2616	26.5	
21-75	1	1173	1174	1283	0.02707	25.6	2.4
21-75	6	1307	1303	1431	0.02716	25.5	3.6
21-76	1	1098	1102	1199	0.02679	25.8	5.5
21-76	6	1224	1215	1338	0.02732	25.3	7.7
		Averages			0.02708	25.5	

TABLE IV.

ESTIMATED U-233 CONTENT

Slug No.	Time	U-233, g
009E6	0	6.1
009E6	247 days	7.5
	Difference	1.4
590D1	0	5.9
590D1	247 days	6.9
	Difference	1.0
21-75	0	6.1
21-75	345 days	6.9
	Difference	0.8
21-76	0	5.5
21-76	345 days	6.3
	Difference	0.8

Note: The difference in U-233 content shown in this table represents the Pa-233 produced in the slug during the irradiation period of this experiment.

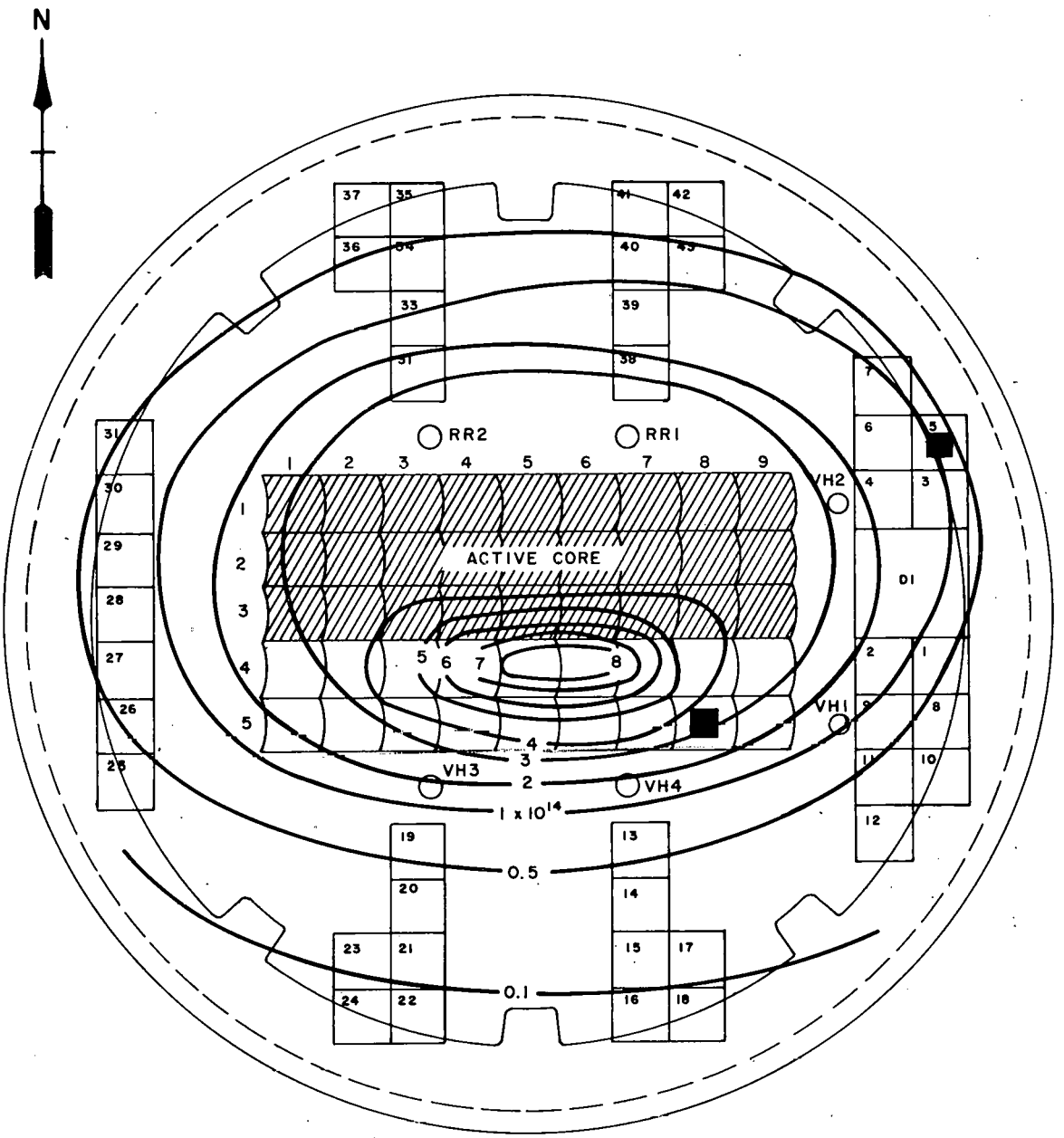

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FIG. 1
MTR THERMAL NEUTRON FLUX AT REACTOR MID-PLANE

■ - LOCATION OF SLUGS IN MTR DURING IRRADIATION

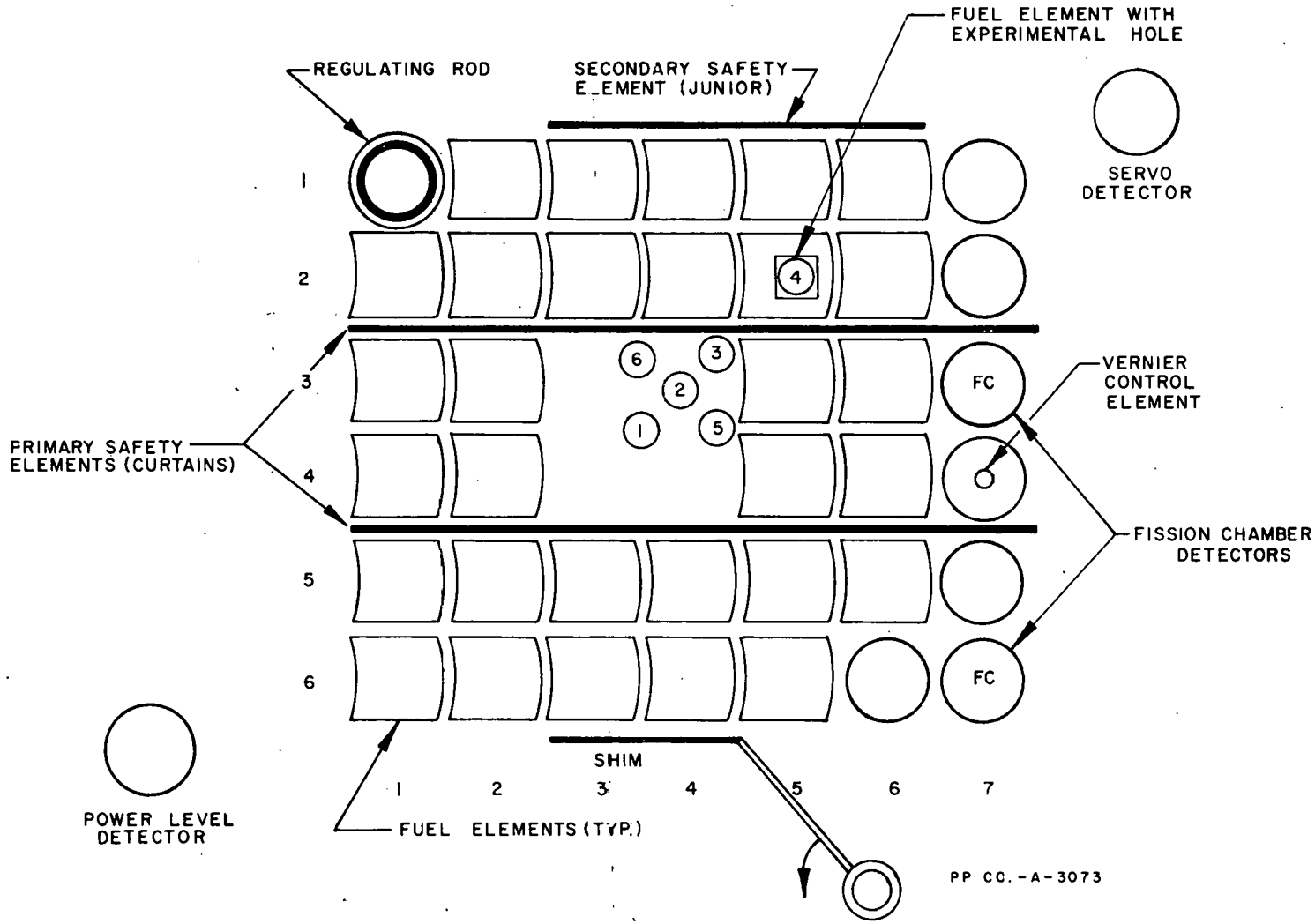

FIG. 2

RMF CORE SHOWING LOCATIONS OF EXPERIMENTAL POSITIONS

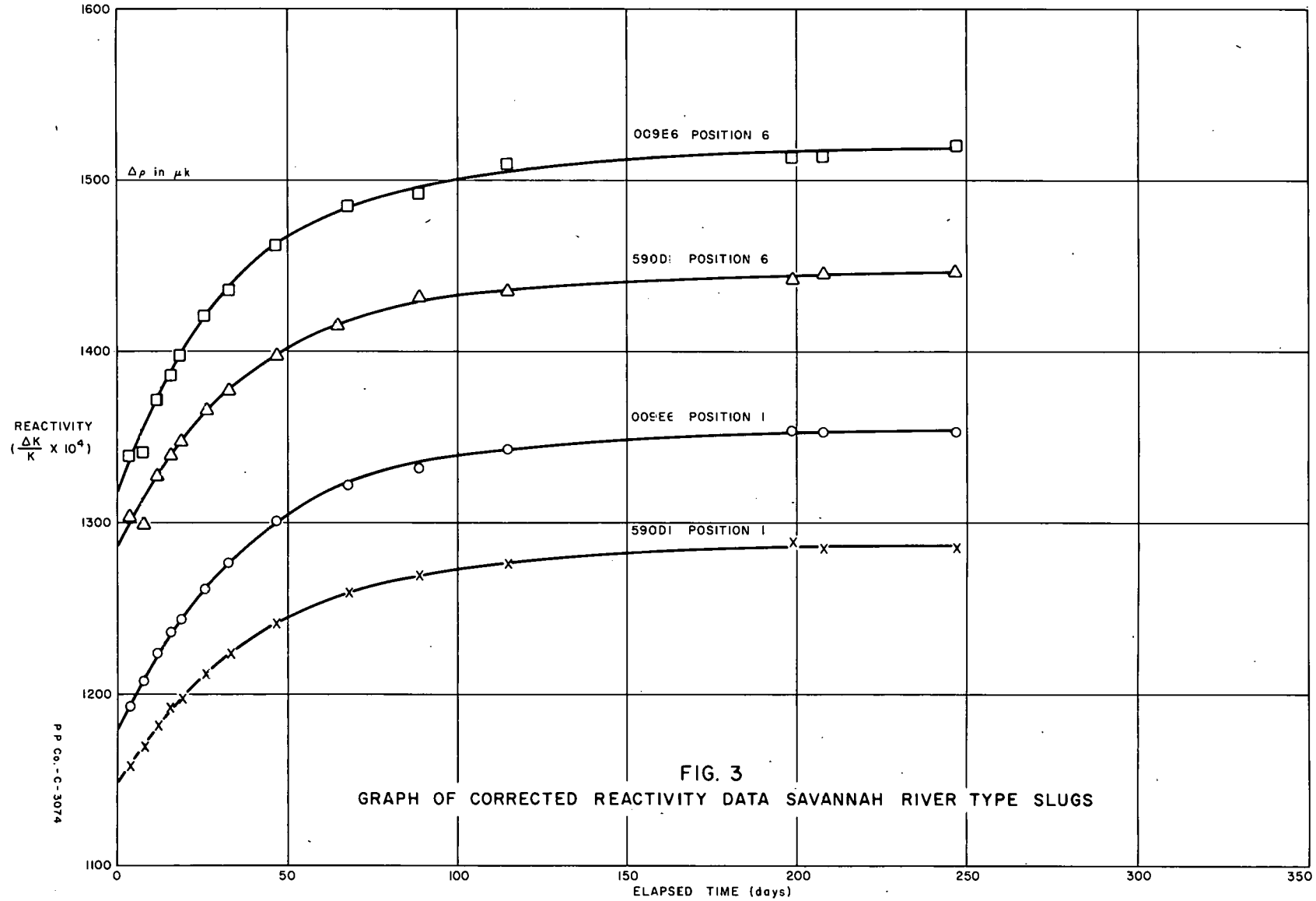

FIG. 3
GRAPH OF CORRECTED REACTIVITY DATA SAVANNAH RIVER TYPE SLUGS

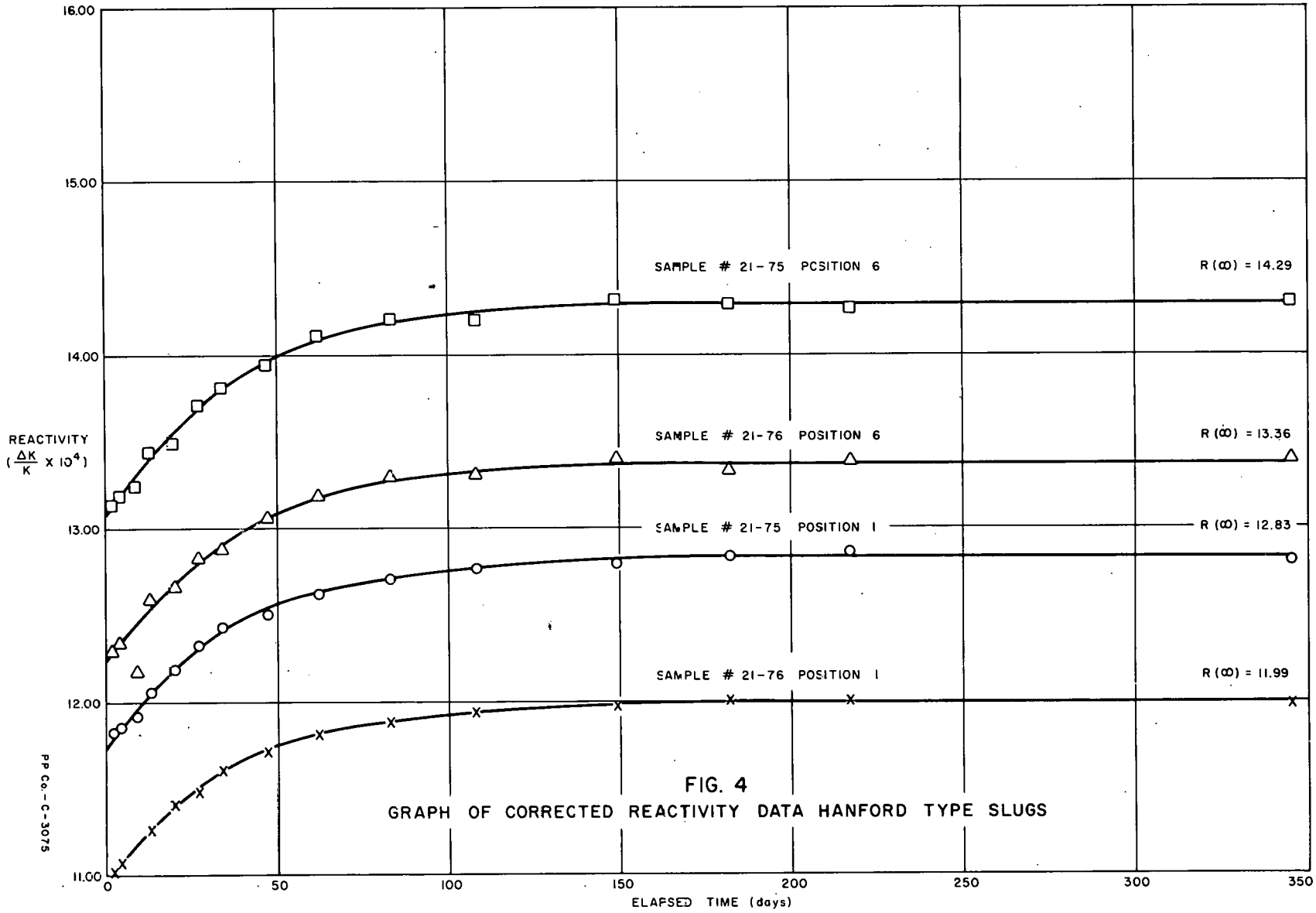

FIG. 4

GRAPH OF CORRECTED REACTIVITY DATA HANFORD TYPE SLUGS

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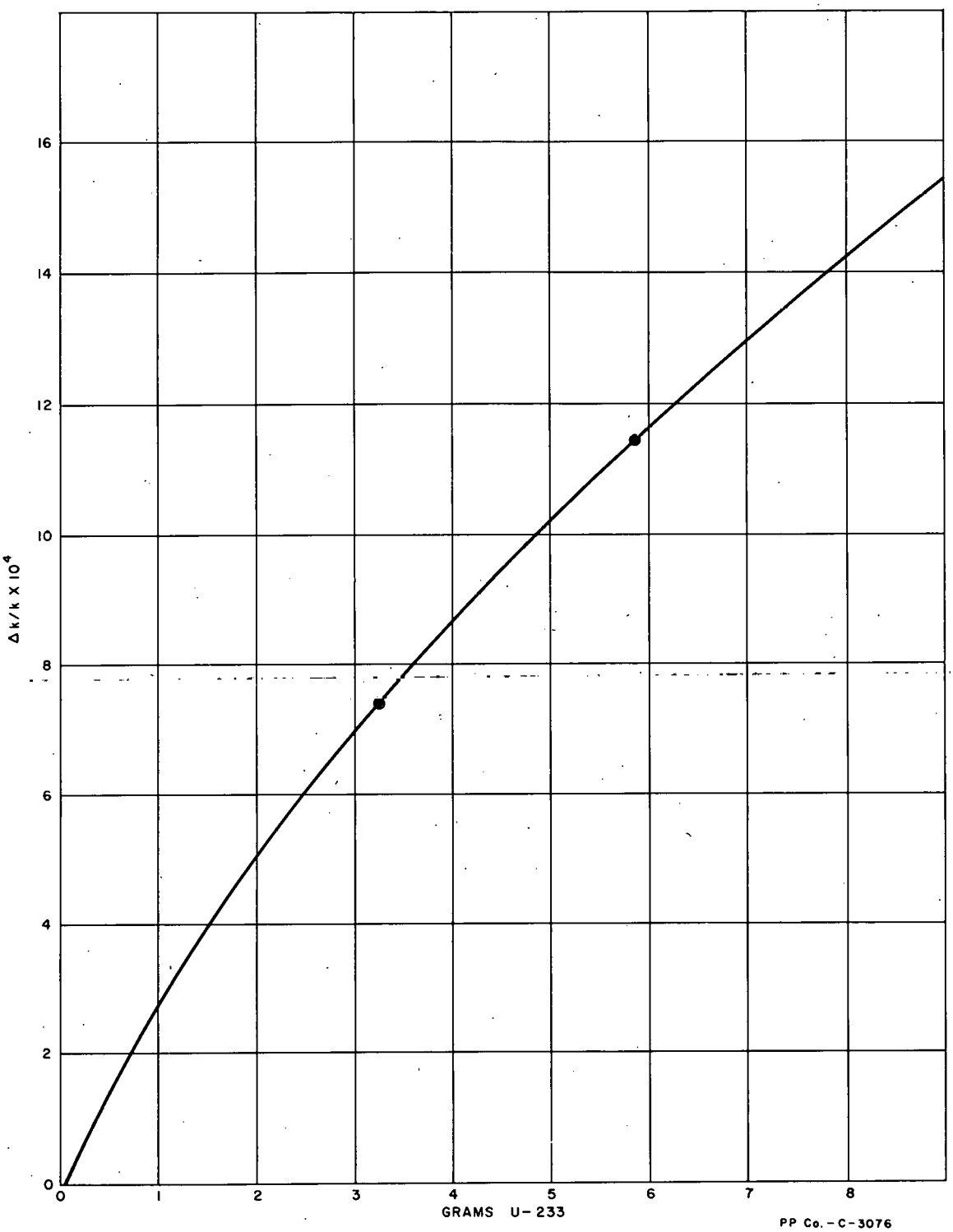

● U-233 CONTENT MEASURED BY CHEMICAL-MASS SPECTROGRAPH METHOD

$U\text{-}233 = 26.15 - 8.33 \ln \left(23.06 - \frac{\Delta k}{k} \times 10^4\right)$

FIG. 5

REACTIVITY U-233 CONTENT

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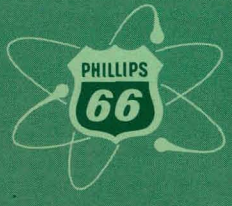

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