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CHEMICAL AND ISOTOPIC ANALYSIS OF IRRADIATED URANIUM SLUGS FROM DEMOUNTABLE SLUG ROD A.G.W. - 1 (P-4498)

CRRP-761 Part B

by

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L.P.V. CORRIVEAU, F. GIRARDI

Chalk River, Ontario

September 1959

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DISCLAIMER

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INTRODUCTION

A number of samples of natural uranium have been irradiated to various levels (0.035 to 0.633 n/kb) in the NRX reactor for studies of the variation of reactivity of natural uranium with neutron irradiation. Measurements of reactivity of the samples before and after irradiation have been made in the GLEEP oscillator at Harwell, and chemical and mass-spectrometric analyses of the samples have been made at Chalk River.

The oscillator results have been presented by Cooper et al. (1), and the present paper presents the results of the chemical and mass-spectrometric analyses. Some of the results have been discussed by Craig et al. (2) at Geneva. Discussion and interpretation of all the results is given in a subsequent paper (3).

EXPERIMENTAL PROCEDURE

The uranium samples, 6 in. x 1.36 in, were cut from standard NRX metal rods and sheathed in aluminum tubing. Cobalt monitors were attached, and the slugs were irradiated near the centre of the NRX lattice. Details of the irradiations and cobalt monitoring are given by Craig et al. (3).

For chemical processing, the slugs were received singly from the NRX underwater storage facility in a 6 in. drawer-type transfer flask, and were transferred from the flask to the chemical cell through the normal sample sleeve. The slugs were then weighed and transferred to the dissolver in a suitable stainless-steel basket. The aluminum sheath was dissolved in 10 liters of 8.8 M sodium hydroxide and both slug and dissolver were washed with distilled water. After the slug had dried it was removed from the dissolver, re-weighed, and its volume was measured by displacement of water. From this weight-volume measurement the slug density was calculated. The slug was then replaced in the dissolver and dissolved in 11 liters of 16 N nitric acid. Acid of this concentration was used to prevent the formation of hexavalent plutonium in the solution. The dissolver solution was now transferred to a measuring tank where its volume was measured to give a preliminary value of the uranium concentration. The solution was then transferred to storage through a 500-ml sample castle. The sample was analysed for total uranium, total plutonium, uranium isotopic composition and plutonium isotopic composition.

ANALYTICAL METHODS

(1) Total Uranium, Gravimetric

Uranium was analysed gravimetrically by precipitation with ammonium hydroxide and ignition to U_3O_8 as described in Scott's Standard Methods of Chemical Analysis, p. 1022, 1024. The method gave an accuracy of ± 0.2% on a 500 mg sample of uranium. Corrections were made for the presence of iron and plutonium in the sample.

(2) Total Uranium, Volumetric

Uranium was analysed volumetrically by reduction to U-IV in a Jones reductor followed by oxidation to U-VI with standard ceric sulphate as described by Fraser (4). The method gives an accuracy of ± 0.3% on a 50 mg sample of uranium. Corrections must be made for the presence of iron and plutonium in the sample.

(3) Total Plutonium, Spectrophotometric

Plutonium was analysed spectrophotometrically using the 602 mμ absorption band of Pu-III. The plutonium was separated from uranium by absorption of the plutonium on an anion-exchange resin then elution with hydroxylamine hydrochloride. The method is described by Allison (5) and gives an accuracy of ± 0.7% on a 2-4 mg sample of plutonium. A correction is usually made, using the zirconium phenyl arsonate - alpha counting method (6), for the amount of plutonium escaping in the column effluent. This correction was in all cases less than 1%.

(4) Total Plutonium, Isotopic Dilution with Plutonium-240

Two aliquots, one containing ∽800 μg of plutonium and the other containing ∽400 μg of plutonium, are removed from the dissolver solution. Only the second aliquot need be measured accurately. To the second aliquot is added ∽400 μg of plutonium containing a high percentage of the heavier isotopes. This aliquot must be accurately measured and the plutonium concentration and isotopic composition must be accurately known. The samples are allowed to stand for 4 or 5 days to allow the spiked sample to reach isotopic equilibrium[☆] and are then carried through the separation procedure (Section 9). The purified plutonium is then mass analysed. From the isotopic compositions of the standard, the unknown and the mixture, and the total plutonium content of the standard, it is possible to calculate the total plutonium concentration in the unknown. The method of calculation and an estimation of errors are given in Appendix 1.

(5) Total Plutonium, Isotopic Dilution with Plutonium-238

Two aliquots, one containing ∽100 μg of plutonium and the other containing ∽50 μg of plutonium, are removed from the dissolver solution. Only the second need be measured accurately. To the second aliquot is added ∽0.19 μg of plutonium-238. This aliquot must be accurately measured and the plutonium-238 disintegration rate of the solution must be accurately known. Both samples are diluted to 15 ml with

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[☆] No detailed study has been made to determine the length of time required to reach isotopic equilibrium but it is known that some considerable time is necessary.

7 M nitric acid and are allowed to stand for 4 or 5 days to allow the spiked sample to reach isotopic equilibrium[✫]. The two samples are now carried through the separation method as described in Section 9 and α-counting discs are prepared from each as described in Section 6. From the ratios of plutonium-238 to plutonium-239 in the spiked and unspiked samples, the total disintegration rate of the standard plutonium-238 solution, and the isotopic composition of unspiked sample it is possible to make an accurate calculation of the total plutonium content of the solution. This calculation, typical spectra, and an estimation of errors are given in Appendix 2.

(6) Total Plutonium, Alpha Counting

In the three most highly irradiated samples, the plutonium-to-uranium ratio was sufficiently high to allow analysis of plutonium by direct alpha counting provided the uranium in the aliquot could be more or less uniformly spread on the disc. This spreading was carried out using tetraethylene glycol (T.E.G.). An aliquot containing about 50 μg of uranium was transferred to a stainless steel source tray in the usual manner. The micropipette used was then washed three times with a solution of T.E.G. in dilute nitric acid (4 drops of T.E.G. in 10 ml of 1 M nitric acid). The solution on the source tray was then evaporated to dryness under an infrared

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[✫] No detailed study has been made to determine the length of time required to reach isotopic equilibrium but it is known that some considerable time is necessary.

lamp and the disc was ignited to redness. The evaporation must take place very slowly in order to avoid decomposition of T.E.G. by hot nitric acid. Two to three hours are necessary for the drying.

The total α-disintegration rate was measured in a Ward-type low-geometry counter (as described in AECL Report CRP-378). The α-spectrum observed in a gridded ion chamber or the low-geometry chamber at about 20 cm pressure showed a prominent group at about 5.15 MeV attributed to Pu-239 and Pu-240, and a weaker group at about 5.5 MeV attributed to Pu-238 and Am-241. The more highly irradiated sample showed a group at about 6.2 MeV attributed to Cm-242 and in the lightly irradiated sample, the two main uranium α-groups at 4.2 and 4.8 MeV were resolved from the tail of the main group. Using the α-spectra, the mass-spectrometer analysis and the specific α-activities of Pu-239 and Pu-240, the amounts of plutonium in the samples were calculated. The method of calculation, a discussion of probable errors and a typical spectrum are given in Appendix 3.

(7) Isotopic Analyses for Uranium

The uranium aliquot from step (c) of the separation procedure (Section 9), containing ~40 mg of uranium, was extracted with 2 ml of 30% TBP in soltrol. The solvent phase was scrubbed 5 times with 0.5 ml portions of 1.0 M hydroxylamine nitrate to remove any residual plutonium.

The uranium was then backwashed with 2 one ml portions of water and the aqueous solution was analysed spectrophotometrically (7). An aliquot containing about 20 mg of uranium was then taken, precipitated with concentrated ammonium hydroxide, washed once with water, and dissolved in 1 ml of 1 M nitric acid. The samples were mass analysed as described by Lounsbury (8). The estimated errors are quoted with the results.

(8) Isotopic Analysis for Plutonium

The plutonium sample from step (g) of the separation procedure (Section 9) was analysed by alpha counting using an estimated specific activity and was mass analysed as described by Lounsbury (9). The estimated errors are quoted with the results.

(9) The Separation Procedure

a) The aliquot, in 7-11 M nitric acid, was passed through a 1 ml column of 250-mesh Dowex-1 in the nitrate form to absorb the anionic plutonium nitrate complex.

b) The uranium and fission products were washed through the column with 20 ml of 7.5 M nitric acid. The strong acid was then displaced with 1 ml of 1 M nitric acid.

c) An aliquot of the effluent containing ∽40 mg of uranium was retained for uranium isotopic analysis and the remainder was discarded.

d) The plutonium was eluted from the column with 4 ml of 1.0 M hydroxylamine nitrate.

e) The plutonium solution was extracted five times with 2 ml portions of 30% TBP-soltrol to remove any uranium left with the plutonium.

f) The plutonium was precipitated from the aqueous phase with concentrated ammonium hydroxide, dissolved in 100 λ of concentrated nitric acid, diluted to 0.5 ml and extracted with two 0.5 ml portions of 30% TBP-soltrol[✫].

g) The plutonium was then backwashed from the organic phase with 3 half-volume 1 M hydroxylamine nitrate washes, precipitated with ammonium hydroxide, washed once with water, and dissolved in 20 λ of concentrated nitric acid. The solution was then diluted to 100 λ with 1 M nitric acid.

RESULTS

A summary of the results obtained is given in the following tables. The starting material for all irradiations was natural uranium with a U-235/U-238 atom ratio (8) of $(0.7257 \pm 0.0007) \times 10^{-2}$. Details of the calibrations, calculations and errors are given in the Appendices.

[✫] The TBP extraction is necessary to separate the plutonium from resin decomposition products.

TABLE I

Uranium Analyses

Slug No.	Nominal Irrad. n/kb	Total U, mg/ml Gravimetric	Total U, mg/ml Volumetric	Atom Ratio U^{235}/U^{238}
8	0.035	241.8 ± 0.5	241.5 ± 0.7	$(0.7116 \pm 0.0010) \times 10^{-2}$
5	0.092	238.0 ± 0.5	238.9 ± 0.7	$(0.6841 \pm 0.0010) \times 10^{-2}$
9	0.227	254.8 ± 0.5	254.7 ± 0.7	$(0.6218 \pm 0.0010) \times 10^{-2}$
11	0.384	242.4 ± 0.5	242.7 ± 0.7	$(0.5671 \pm 0.0016) \times 10^{-2}$
4	0.633	277.2 ± 0.5	277.7 ± 0.7	$(0.4764 \pm 0.0012) \times 10^{-2}$

TABLE II

Total Plutonium Analyses

Slug No.	Total Pu mg/ml Spectrophotometric	Total Pu mg/ml Alpha Counting	Total Pu mg/ml Pu^{240} Dilution	Total Pu mg/ml Pu^{238} Dilution
8	-	-	0.0316 ± 0.00035	0.0304 ± 0.0002
5	0.0806 ± 0.0006	-	0.0797 ± 0.0008	0.0773 ± 0.0005
9	0.195 ± 0.0014	0.194 ± 0.0014	(0.199 ± 0.002)	0.195 ± 0.002
11	0.295 ± 0.002	0.293 ± 0.002	(0.303 ± 0.003)	0.301 ± 0.003
4	0.505 ± 0.004	0.498 ± 0.003	0.513 ± 0.007	0.514 ± 0.005

( ) Values must be considered suspect as isotopic equilibrium not certain.

TABLE III

Plutonium Isotopic Analysis[*]

Slug No.	Atom % Pu-239	Atom % Pu-240	Atom % Pu-241	Atom % Pu-242	Date of Analysis
8	99.268 ± 0.008	0.727 ± 0.006	0.005 ± 0.005	-	Aug. 1/58
5	98.146 ± 0.017	1.824 ± 0.016	0.030 ± 0.007	-	July 4/58
9	95.209 ± 0.012	4.537 ± 0.011	0.254 ± 0.005	-	Apr. 17/58
11	92.241 ± 0.018	7.163 ± 0.016	0.596 ± 0.009	-	March 25/58
4	87.295 ± 0.052	11.267 ± 0.051	1.320 ± 0.010	0.118 ± 0.005	Mar. 19/58

[*] The results have not been corrected for the decay of Pu-241.

TABLE IV

Plutonium Isotopic Analysis[*]

Slug No.	Weight % Pu-239	Weight % Pu-240	Weight % Pu-241	Weight % Pu-242
8	99.265 ± 0.008	0.730 ± 0.006	0.005 ± 0.005	-
5	98.138 ± 0.017	1.832 ± 0.016	0.030 ± 0.007	-
9	95.189 ± 0.012	4.555 ± 0.011	0.256 ± 0.005	-
11	92.209 ± 0.018	7.190 ± 0.016	0.601 ± 0.009	-
4	87.243 ± 0.052	11.307 ± 0.051	1.330 ± 0.010	0.120 ± 0.005

[*] The results have not been corrected for the decay of Pu-241.

TABLE V

Density of Irradiated Slugs

Slug No.	Weight of Slug g	Volume of Slug cm^3	Density g/cm^3
8	-	-	-
5	-	-	-
9	2705	143.6	18.84
11	2704	143.2	18.88
4	2703	150.8	17.93 ✮

✮ Slug No. 4 had a pronounced conical swelling at one end.

DISCUSSION

For each slug measured there are two to four values for the total plutonium content. There are two possible ways to obtain a "best value." One is to weigh each value according to the quoted standard error, i.e. the weight, w, is equal to $\frac{1}{(\text{error})^2}$. These weight averages are given in Table VI. For each value it is possible to calculate two errors, $\alpha_s = \frac{1}{\sqrt{\Sigma w}}$, based entirely on the quoted errors, or $\alpha d = \sqrt{\frac{\Sigma w_i \, d_i^2}{(n-1)\Sigma w_i}}$, where d_i is the deviation of each value from the weighted average, w its weight, and n the number of values averaged. For the spread in the results to be consistent with the quoted errors, $\alpha_s \backsimeq \alpha_d$. The values of α_d and $\frac{\alpha_d}{\alpha_s}$ are given in Table VI. The average value of α_d/α_s is 2.4 showing that the deviations are larger than expected for the quoted errors.

Another way to obtain a "best value" is to disregard the quoted errors and take an average. These values are also given in Table VI along with the corresponding values for α_d.

The "best values" are probably the weighted averages with the larger of α_s or α_d as the error.

TABLE VI

Total Plutonium Analyses - Mean Values

Slug No.	Weighted Average[*] mg/ml	α_s	α_d	$\frac{\alpha_d}{\alpha_s}$	Unweighted Average[*] mg/ml	α_d
8	0.0307	0.00017	0.0005	3	0.0310	0.0007
9	0.0789	0.0004	0.0016	4	0.0792	0.0010
5	0.195	0.009	0.0003	0.3	0.195	0.004
11	0.295	0.0013	0.002	1.5	0.296	0.0017
4	0.504	0.002	0.004	2	0.508	0.004

[*] The bracketed results for slugs 9 and 11 (Table II) have been excluded from these averages.

ACKNOWLEDGMENT

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APPENDIX I

Calculation of Total Plutonium Concentration in Pu-240 Dilution Analysis

The spiking solution, F.P.-49-335, was analysed spectrophotometrically for total plutonium content, mass spectrometrically for plutonium isotopic composition and spectrographically for impurities, giving the following results.

Total Pu - 93.6, 93.6, 93.7, 93.8 mg/g

✱ Atom % Pu-239 - 60.962 ± 0.044 ✱Wt. % Pu-239 = 60.845
" " Pu-240 - 33.333 ± 0.040 " " Pu-240 = 33.408
" " Pu-241 - 4.611 ± 0.015 " " Pu-241 = 4.640
" " Pu-242 - 1.094 ± 0.010 " " Pu-242 = 1.107

Impurities (%) - Ba <0.02, Be <0.02, Ca >0.5, Cr <0.06, Cu <0.06, Fe <0.75, K 0.6, Mg <0.02, Na 0.05, Ni <0.06, Pb 0.10, Si 0.15, Sr <0.02, Ti <0.02, U <0.6, Zn >0.5, Zr <0.02.

A 0.544-g aliquot from F.P.-49-335 was diluted to 25 ml with 7 N HNO_3 to give a plutonium concentration of 2.039 mg/ml in the actual solution used for spiking. This solution was checked spectrophotometrically and gave a concentration of 2.05 mg/ml. It should be pointed out that cells with a 1-cm light path were used in the analysis of the concentrated solution whereas cells with a 5 cm light path were used in the analysis of the dilute solution. Both sets of cells were calibrated against an accurately weighed sample of plutonium metal of known purity.

✱ The mass spectrometer analysis were made on Feb. 26/58.

The weight per cent of Pu-239 in the spiked sample may be related to the weight percentages of Pu-239 in the unspiked sample and spiking solution by the relation

$$m_s\, P_s + m_u\, P_u = P_m\, (m_s \pm m_u)$$

where m_s = the mass of spiking plutonium added,

m_u = the mass of unspiked plutonium in the volume, V_u, of unknown added,

P_s = the weight per cent Pu-239 in the spiking plutonium,

P_u = the weight per cent Pu-239 in the unspiked plutonium,

P_m = the weight per cent Pu-239 in the spiked plutonium.

$$m_u = m_s \left\{\frac{(Pm-Ps)}{(P_u-P_m)}\right\}$$

The plutonium concentration is then $\frac{m_u}{V_u}$ in any consistent units.

Estimation of Errors in Pu-240 Dilution Analysis

Although the four plutonium values on the plutonium-240 spiking solution deviate only 0.1% from the mean, a small error is possible in dilution so a probable error of ± 0.2% has been placed in the plutonium content of the spiking solution.

Micro pipettes in the 50, 100, 200 and 500 λ range have been calibrated with mercury and have been found to be consistently accurate to ± 0.2%. The U.S. Bureau of Standards requires that macro pipettes conform to the following specifications: 1 ml - 0.6%, 2 ml - 0.3%, 5 and 10 ml - 0.2%. These errors have been placed on the measurement of the standard and unknown solutions.

Errors in the mass-spectrometer measurements must be imposed upon the differences $P_m - P_s$ and $P_u - P_m$. Thus the percentage error in $P_m - P_s$ and $P_u - P_m$ is determined from

$$\frac{E_{P_m}}{P_m-P_s} \times 100, \quad \frac{E_{P_s}}{P_m-P_s} \times 100, \quad \frac{E_{P_u}}{P_u-P_m} \times 100 \quad \text{and} \quad \frac{E_{P_m}}{P_u-P_m} \times 100,$$

where the E's represent the respective mass-spectrometer deviations.

The total probable error is determined by taking the square root of the sum of the squares of the individual errors.

APPENDIX 2

Calculation of Total Plutonium Concentration in Pu-238 Dilution Analysis

The spiking solution was prepared by irradiating americium-241 to form americium-242 and separating plutonium-238 from the decay chain by ion exchange. Alpha-spectral analysis showed that it contained none of the other isotopes of plutonium.

The dpm due to Pu-239 and Pu-240 in the volume V_u of unknown sample used can be determined from the equation

$$D_u = \frac{D_s}{R_m - R_u}$$

where D_u = the dpm Pu-239 + Pu-240 in the unspiked sample,

D_s = the dpm Pu-238 added as spike,

R_m = the ratio of the Pu-238 peak to the Pu-239 + Pu-240 peak in the spiked sample,

R_u = the ratio of the Pu-238 peak to the Pu-239 + Pu-240 peak in the unspiked sample.

But $D_u = N_9\lambda_9 + N_o\lambda_o$

where N_9 = the number of atoms of Pu-239 in the volume V_u,

N_o = the number of atoms of Pu-240 in the volume V_u,

λ_9 = the disintegration constant of Pu-239 (5,400 x 10^{-11} min.$^{-1}$ corresponding to a half life of 24,400 years (±0.3%)).

λ_o = the disintegration constant of Pu-240 (1.973 x 10^{-10} min.$^{-1}$ corresponding to a half life of 6,580 years) (10).

Then
$$N_9 = \frac{D_s}{(R_m - R_s)\,\lambda_9\,(1 + \frac{N_o\lambda_o}{N_9\lambda_9})}$$

$\frac{N_o}{N_9}$ can be obtained from mass-spectrometer analysis.

Converting from atoms, N_9, to milligrams, M_9, gives

$$M_9 = \frac{3.968 \times 10^{-19}\,D_s}{(R_m - R_s)\,\lambda_9\,(1 + \frac{N_o\lambda_o}{N_9\lambda_9})}$$

The total plutonium concentration, M_T, in milligrams per unit volume of solution is then given by

$$M_T = \frac{3.968 \times 10^{-19}\,D_s}{\lambda_9\,m_9\,V_u\,(R_m - R_s)\,(1 + \frac{N_o\lambda_o}{N_9\lambda_9})}$$

where m_9 is the mass fraction of plutonium-239 from mass-spectrometer analysis.

Simplifying the above equation gives

$$M_T = \frac{7.348 \times 10^{-9}\,D_s}{M_9\,V_u\,(R_m - R_s)\,(1 + 3.653\,\frac{N_o}{N_9})}$$

In all plutonium samples containing Pu-241, the Am-241 formed from the β-decay of the Pu-241 grows in with a 13.0 year half-life. The Am-241 α-group was unresolved from the Pu-238 α-group in these measurements. Therefore if R_m and R_u were measured at different times relative to the plutonium separation, a correction should be applied for this activity. This correction was estimated from the Pu-241 content (given in the mass analysis) and the relevant decay constants, and was found to be less than 0.1% in all cases.

Estimation of Errors in Pu-238 Dilution Analysis

The Pu-238 spiking solution was analysed by alpha counting in a Ward-type low-geometry chamber. The accuracy of this analysis was estimated to be ± 0.5%.

The Pu-239 + Pu-240 and Pu-238 α-groups were not completely resolved in these measurements so that the tail from the Pu-238 group must be subtracted from the Pu-239 + Pu-240 group. The error in this subtraction was estimated to be about 20% of the quantity subtracted and was usually about 0.5%.

Pipetting errors for each of the three pipettings required were estimated at ± 0.2%.

The error attributed to the disintegration constant of Pu-239 was ± 0.3%. That contributed by the error in λ_0/λ_9 was negligible.

The total probable error was obtained as in Appendix 1 by taking the square root of the sums of the squares of the individual errors.

APPENDIX 3

Calculation of Total Plutonium Concentration in α Analysis

The total α-disintegration rate A given by a sample of volume V_u may be written as

$$A = N_u\lambda_u + N_9\lambda_9 + N_o\lambda_o + N_8\lambda_8 + N_1\lambda_1 + N_2\lambda_2 \qquad [1]$$

where N_u, N_9, N_8, N_o, N_1, N_2 are the number of atoms of uranium, Pu-239, Pu-240, Pu-238, Am-241 Cm-242, respectively, and λ_u, λ_9, λ_8, λ_o, λ_1, λ_2, are the disintegration constants. λ_u, λ_9, and λ_o have the values 5.93×10^{-16}, 5.400×10^{-11}, and 1.973×10^{-10} min.^{-1} respectively. λ_8, λ_1 and λ_2 need not be known. Rearranging equation [1] gives

$$N_9 = \frac{A - N_u\lambda_u}{\lambda_9\left(1+\frac{N_o\lambda_o}{N_9\lambda_9}\right)\left(1 + \frac{N_8\lambda_8 + N_1\lambda_1}{N_9\lambda_9 \quad N_o\lambda_o} + \frac{N_2\lambda_2}{N_9\lambda_9 + N_o\lambda_o}\right)} \qquad [2]$$

N_u can be obtained from uranium analysis (Table 1), N_o/N_9 from mass-spectrometer analysis, $\frac{N_8\lambda_8 + N_1\lambda_1}{N_9\lambda_9 + N_o\lambda_o}$ and $\frac{N_2\lambda_2}{N_9\lambda_9 + N_o\lambda_o}$ from the α spectra observed in the gridded ion chamber or low-geometry chamber at high pressure (20 cm.). Converting atoms, N_9, to milligrams, M_9, gives

$$M_9 = 3.968 \times 10^{-19} N_9 \qquad [3]$$

The total plutonium concentration, M_T, in milligrams per unit volume of solution is then given by,

$$M_T = \frac{M_9}{V_u m_9} = \frac{7.348 \times 10^{-9} (A - N_u\lambda_u)}{V_u m_9 \left(1 + \frac{N_o\lambda_o}{N_9\lambda_9}\right)\left(1+\frac{N_8\lambda_8 + N_1\lambda_1}{N_9\lambda_9 \quad N_o\lambda_o} + \frac{N_2\lambda_2}{N_9\lambda_9 + N_o\lambda_o}\right)}$$

where m_9 is the mass fraction of plutonium-239 from mass-spectrometer analysis.

Estimation of Errors in Total Plutonium Analysis by α-Counting

The low geometry chamber efficiency, calculated for a point source, was 1 in 703.7 ± 0.1%. The correction for the source size, 0.87 inches, taken from CRP-378, is 0.58% giving 1 in 707.8 ± 0.1%. This error includes only the error in the measurement of aperture size and distance from the source. Another possible error arises from possible non-uniform spreading of the sample material; for instance, if it were all piled at the centre the error would be 0.58%. A reasonable estimate is probably ±0.2% for this effect.

Thick-source losses were investigated by comparing the specific activities observed for samples of different sizes. In a preliminary experiment, evaporated samples of about 50 and 100 μg of uranium were found to have up to 5% difference in specific activity. This was attributed to thick-source losses caused by a non-uniform spreading of the uranium. Subsequent samples of 25, 50 and 100 μg were prepared with a spreading agent. Again the specific activity of some of the 100 μg samples were lower than that of the 25 and 50-μg samples, indicating as much as 4% thick-source loss. The specific activity of the 20 and 50-μg samples were the same within the counting statistics of about 1% and were averaged for the final result.

Because of the source-size limitation, the counting rates were low, 4.5 - 60 counts/min., and the chamber background, 0.10 ± 0.01 counts/min., was significant, but the correction introduced less than 0.2% error.

The error contributed by uncertainties in the half lives of plutonium-239 and plutonium-240 is estimated at 0.4%.

The error in each of two pipettings is 0.2%.

The total probable error is taken as the square root of the sum of the squares of the individual errors.

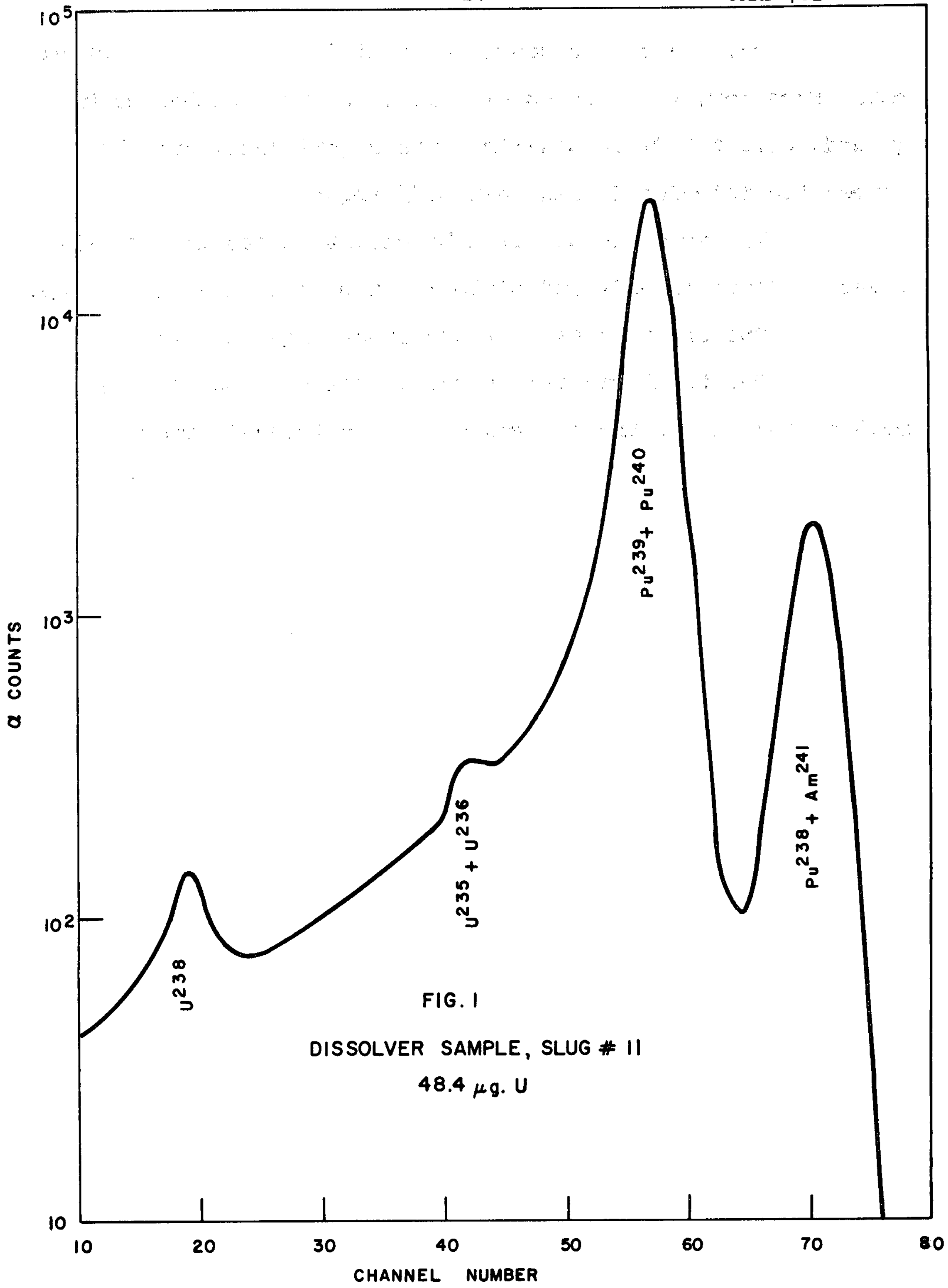

FIG. 1

DISSOLVER SAMPLE, SLUG # 11

48.4 μg. U

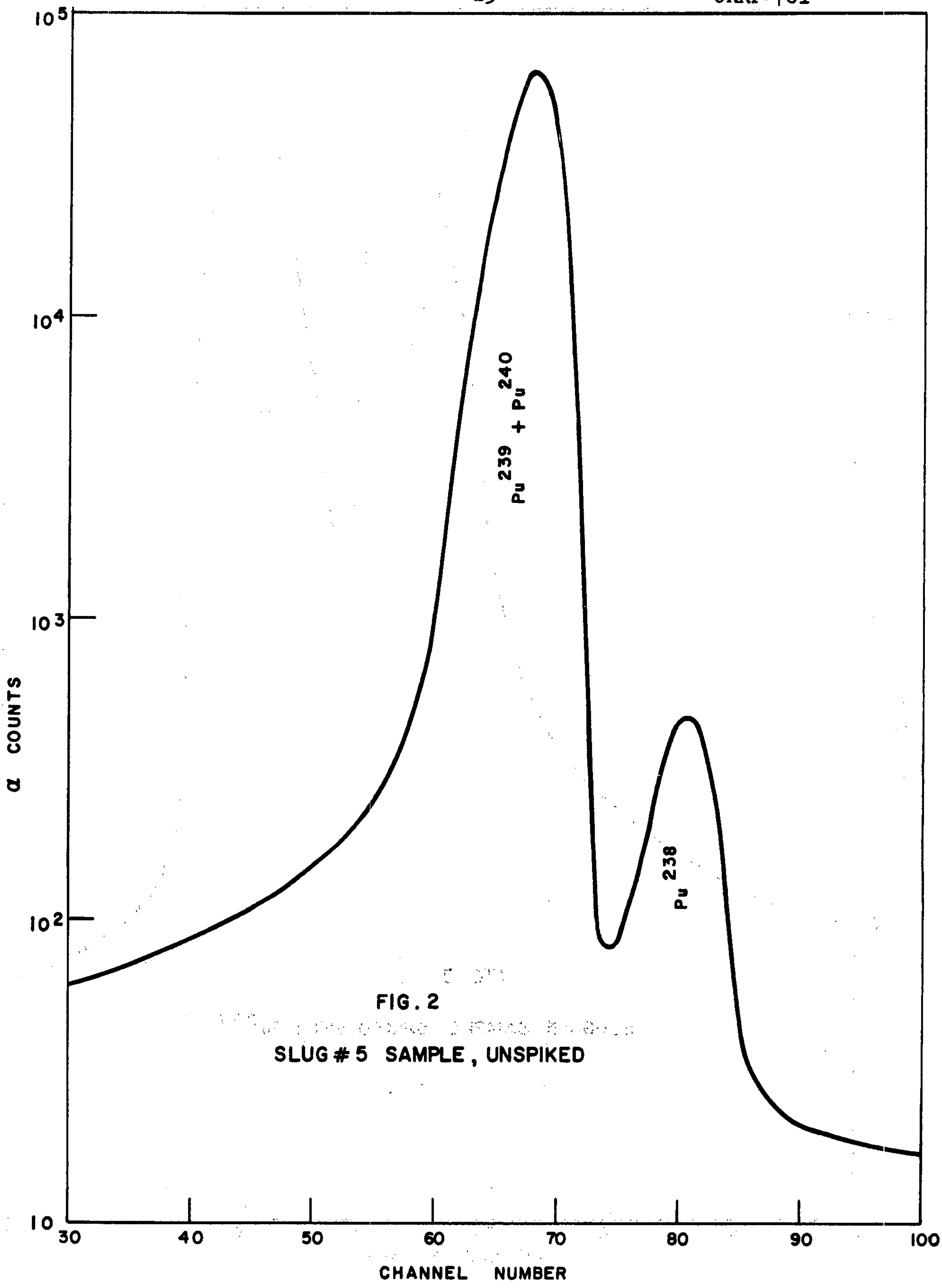

FIG. 2

SLUG # 5 SAMPLE, UNSPIKED

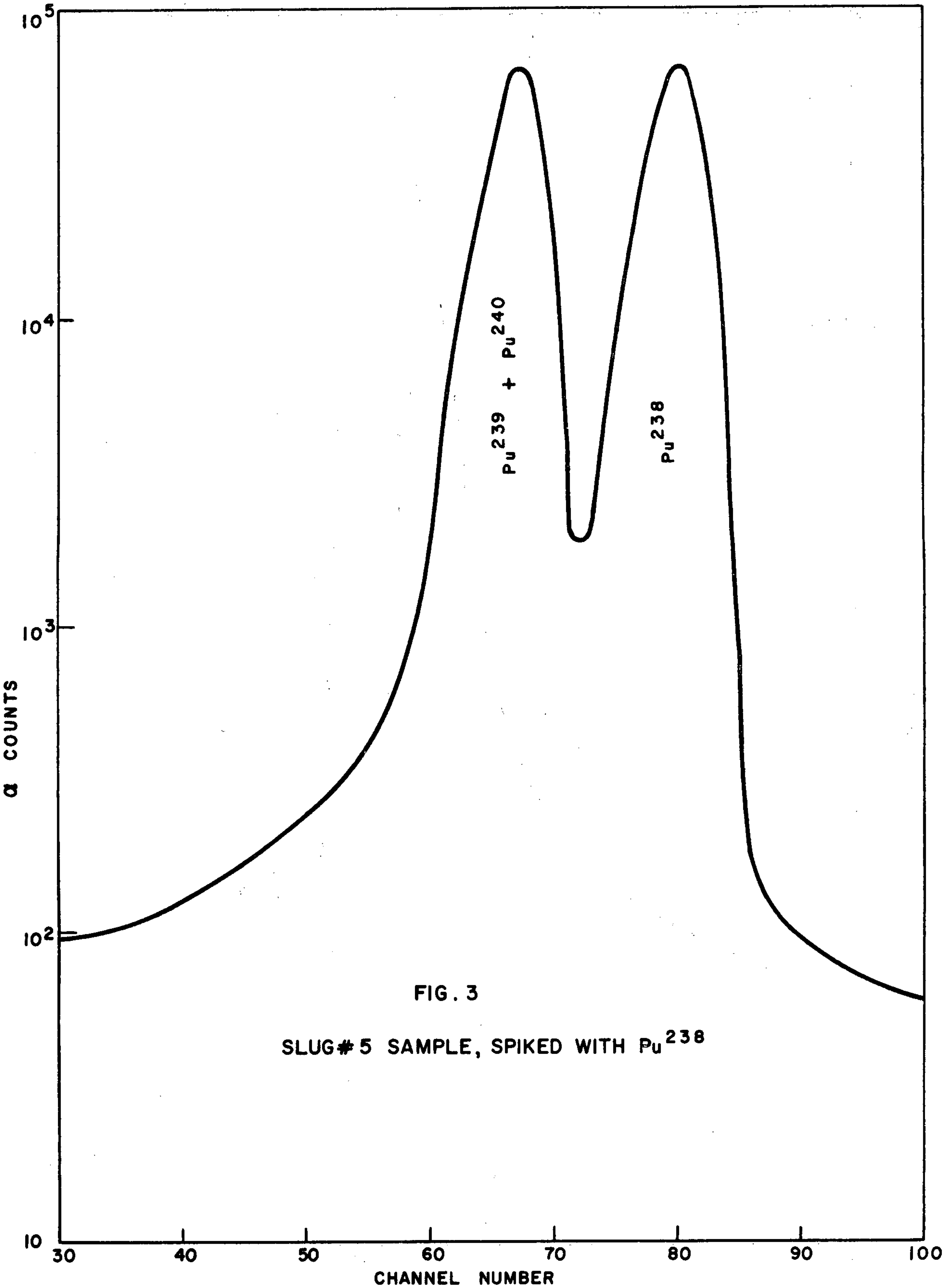

FIG. 3

SLUG# 5 SAMPLE, SPIKED WITH Pu^{238}