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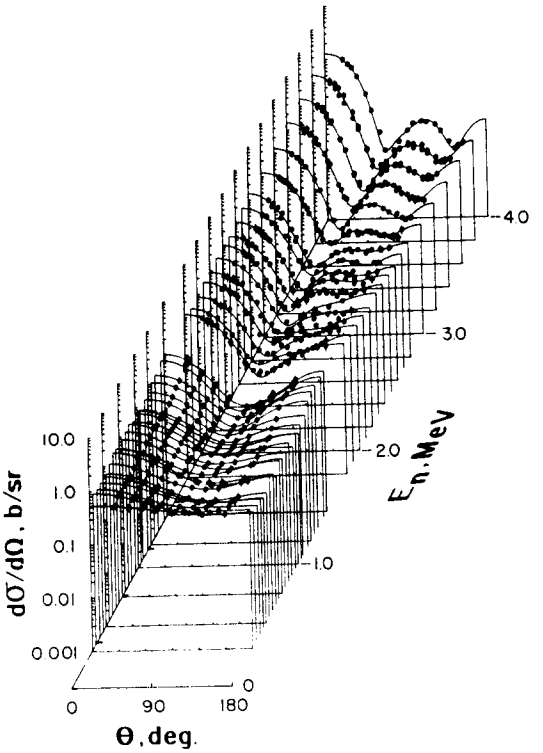
ANL/NDM-38

THE ALPHA AND SPONTANEOUS FISSION HALF-LIVES OF ^{242}Pu

by

J. W. Meadows

December 1977



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In October 1977, the U. S. Energy Research and Development Agency (ERDA) was incorporated into the U. S. Department of Energy. The research and development functions of the former U. S. Atomic Energy Commission had previously been incorporated into ERDA in January 1975.

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NUCLEAR DATA AND MEASUREMENTS SERIES

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THE ALPHA AND SPONTANEOUS FISSION HALF-LIVES OF ^{242}Pu*

by

J. W. Meadows
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ABSTRACT

The alpha and spontaneous fission half-lives of ^{242}Pu are measured relative to the alpha half-life of ^{239}Pu. If the alpha half-life of ^{239}Pu is 24290 ± 70 y, the ^{242}Pu alpha half-life is (3.736 ± .029) x 10^5 y and the spontaneous fission half-life is (6.79 ± .04) x 10^{10} y.

*This work supported by the U. S. Department of Energy.

I. INTRODUCTION

The early measurements of the ^{242}Pu alpha half-life by Thompson et al[1] and by Asaro[2] gave 5 x 10^5 y and 9 x 10^5 y respectively. Later measurements[3-8] with better accuracies clustered around 3.75 x 10^5 y. However, even the more recent values[6-8] differ by amounts that are large compared to their assigned errors. There are only a few measurements of the spontaneous fission half-life[3,5,9,10] and the extreme values differ by nearly 15%.

In the course of a program of fission cross section ratio measurements a group of samples were prepared from high purity ^{242}Pu and from mixtures of ∿90% ^{242}Pu and ∿9% ^{239}Pu. This provided an opportunity to compare the ^{242}Pu alpha and spontaneous fission half-lives to the alpha half-life of ^{239}Pu. Since the ^{239}Pu alpha half-life is well known[11] this method should be capable of yielding accurate values for the ^{242}Pu half-lives. The conditions for the measurement were good. The isotopic composition of these samples was well determined, the alpha activities of the ^{242}Pu and ^{239}Pu in the mixed samples were roughly equal, and their alpha decay energies were sufficiently different so the two alpha groups could be easily separated. This report contains the results of that comparison.

II. EXPERIMENTAL PROCEDURES

High purity ^{242}Pu and ^{239}Pu were obtained from the Research Materials Collection at Oak Ridge National Laboratory. The isotopic analysis of the ^{242}Pu used in this measurement was that supplied with the material and is the same as that given for sample 42 in Table 1. The ^{239}Pu used to prepare the mixed samples was 99.998% ^{239}Pu.

The samples were prepared by electro-deposition of the plutonium onto molybdenum plates followed by conversion to the oxide. The deposits were 2.5 cm in diameter and had area densities ranging from ∿40 to ∿70 μg Pu/cm^2. The

mixed samples were analyzed for isotopic composition after completion of the measurements. The results are listed in Table 1.

The samples were alpha counted in a low geometry counter with a geometry factor of 939 ± 3. A typical alpha spectrum is shown in Fig. 1. Although the resolution of the surface barrier detector used in the counter was 22 keV the principal components of the ^{242}Pu alpha peak cannot be resolved due to the thickness of the deposits. However, separation of the ^{242}Pu and ^{239}Pu alpha groups is good and the correction for the tail of the ^{239}Pu peak that extends under the ^{242}Pu peak is small.

The spontaneous fissions were counted in an ionization chamber of the type previously described.[12] A fission spectrum is shown in Fig. 2. The electrode separation of the ion chamber is much less than the fission fragment range so the usual two-humped fission fragment spectrum is not observed. However there is good separation between the alphas and the fissions. The number of fissions lost below the discriminator bias level was estimated by linear extrapolation to zero pulse height. The correction factor for the fissions not observed due to the finite deposit thickness was estimated by

$$S = 1 + T/(2r) \qquad (1)$$

where T is the deposit thickness in terms of mg Pu/cm^2 and r is the average range of the fission fragments in the deposit expressed as mg Pu/cm^2. Since the deposit is an oxide the value of r is considerably less than the range in the metal. The value used, 4.7 mg/cm^2, was actually obtained from measurements on a series of uranium oxide deposits. The value for plutonium oxide should be a little different but since the loss is only ∿0.5% the use of the uranium value introduces no significant error.

III. RESULTS

A. The Alpha Half-life

Inspection of Fig. 1 shows that the alpha spectrum consists of three well separated peaks, so determining the relative peak areas is fairly straightforward. The tail of the central peak does extend under the low energy peak but the correction is small. It is largest for the ^{242}Pu + ^{239}Pu samples and also depends on the deposit thickness. For these samples it was always <1%.

The high energy peak is almost entirely due to ^{238}Pu. It also contains any alphas from ^{241}Am formed by β-decay of ^{241}Pu. The low energy peak and the middle peak are almost entirely due to ^{242}Pu and ^{239}Pu respectively but there is some interference from other plutonium isotopes. Some of the very weak ^{239}Pu alpha groups fall into the ^{242}Pu peak but they only amount to 0.08%. The ^{241}Pu alphas also fall into this peak but the concentration is low and the alpha half-life is long so this contribution is only 0.02%. The ^{240}Pu alphas fall into the middle peak. This is the most important interference and amounts to ∿4%.

When all the interferences are considered explicitly the ^{242}Pu alpha half-life is

$$t_\alpha(242) = RF(242)\{[P(1 + R) - R]F(239)/t_\alpha(239) + F(240)/t_\alpha(240) - RF(241)/t_\alpha(241)\}^{-1} \quad (2)$$

where R is the ratio of the area of the middle peak to the low energy peak, F is the mole fraction of the designated isotope, t_α is its alpha half-life and P is the fraction of ^{239}Pu alphas falling into the middle peak. For this sample P is 0.9992. The half-lives used are listed in Table 2. The value for the ^{239}Pu half-life is the one recommended by Lemmel.[11] A recent measurement by Jaffey et al[16] is in good agreement. Values of R for samples 44 and 50 were based on the average of several measurements. The results for the ^{242}Pu half-life were

Sample 44	$(3.757 \pm .040) \times 10^5$ y.
Sample 50	$(3.715 \pm .028) \times 10^5$ y.
Unweighted Av.	$(3.736 \pm .029) \times 10^5$ y.

The principal sources of error are listed in Table 3. The error in R is based on the scatter of a number of determinations. It is larger than the statistical error and is the principal uncertainty in the measurement.

B. The Spontaneous Fission Half-life

The measurement of the spontaneous fission half-life, t_{SF}, is essentially a measurement of the spontaneous fission decay rate relative to the alpha decay rate as shown in the following equation.

$$t_{SF}(242) = t_{\alpha}(242) \frac{G(C_{\alpha} - B_{\alpha})}{(C_{SF} + D - B_{SF})S}$$

where C_{α} is the count rate in the low energy alpha peak, G is the alpha counter geometry factor, B_{α} is the count rate in the low energy peak due to other isotopes, C_{SF} is the spontaneous fission count rate, B_{SF} is the number of spontaneous fissions due to other isotopes, D is the correction for counts below the discriminator level and S is the factor used to correct for losses in the plutonium deposit. Spontaneous fission rates were measured for samples 42 and 49. There was no isotopic analysis for sample 49 but it was known to be similar to that of samples 44 and 50. This is confirmed by the alpha spectrum. In any case, once the ^{242}Pu alpha half-life is known, the isotopic analysis is needed only to calculate the minor corrections B_{α} and B_{SF} which were less than 0.1%.

The results for the ^{242}Pu spontaneous fission half-life was

Sample 42	$(6.78 \pm .06) \times 10^{10}$ y.
Sample 49	$(6.80 \pm .06) \times 10^{10}$ y.
Unweighted Av.	$(6.79 \pm .05) \times 10^{10}$ y.

The principal sources of error are listed in Table 3.

IV. COMMENTS

The present values of the ^{242}Pu alpha and spontaneous fission half-lives are compared with other measurements[3-10] in Table 4. The early results[1,2] are not included since their errors are large. The values of the ^{242}Pu half-lives which were measured relative to the half-lives of other plutonium isotopes were adjusted by means of the appropriate half-life given in Table II. The errors are those given by the original authors.

The only other measurement made relative to ^{239}Pu gave (3.851 ± .016) x 10^5 y[6] which is in rather poor agreement with the present value of (3.736 ± .025) x 10^5 y. However the present value is consistent with the recent calorimetric value of (3.763 ± .009) x 10^5 y and also with the unweighted mean of all values ((3.759 ± .033) x 10^5 y). The unweighted mean of all values of the spontaneous fission half-life is (6.86 ± .18) x 10^{10} y while the weighted mean is (6.78 ± .04) x 10^{10} y. Either of these is in good agreement with the present value of (6.79 ± .05) x 10^{10} y.

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TABLE 1. The Isotopic Composition of the Samples in Mole %

Isotope	Sample 42	Sample 44	Sample 50
^{238}Pu	0.006%	0.007%	0.006%
^{239}Pu	0.008	9.174	9.250
^{240}Pu	0.107	0.097	0.102
^{241}Pu	0.137	0.099	0.097
^{242}Pu	99.742	90.622	90.546

TABLE 2. The Alpha and Spontaneous Fission Half-lives of the Plutonium Isotopes

Isotope	Half-Life: Alpha	Half-Life: Spontaneous Fission
^{238}Pu	87.71 ± .03 y[a]	5.2×10^{10} y[d]
^{239}Pu	24290 ± 70 y[b]	5.4×10^{14} y[d]
^{240}Pu	6537 ± 10 y[c]	1.33×10^{11} y[d]
^{241}Pu	6.04×10^{5} y[d]	
^{242}Pu	$(3.736 \pm 0.25) \times 10^{5}$ y[e]	$(6.79 \pm .05) \times 10^{10}$ y[e]

[a]Ref. 13.
[b]Ref. 11.
[c]Ref. 14.
[d]Ref. 15.
[e]This Experiment.

TABLE 3. The Error in the Final Result Due to Uncertainties in the Quantities in Eqs. (2) and (3)

Quantity	% Error in Half-life
Alpha Half-life	
R	0.5-1.0
F(242)	0.1
F(239)	0.2
F(240)	0.2
$t_\alpha(239)$	0.3
Spontaneous Fission Half-life	
C_α	0.3
C_{SF}	0.4-0.5
G	0.3
D	0.2
S	0.1
$t_\alpha(242)$	0.67

TABLE 4. Comparison of ^{242}Pu Alpha and Spontaneous Fission Half-lives

Reference	Method	Adjusted Half-life[a]	
		Alpha	Spontaneous Fission
3	Relative to ^{238}Pu t_α	$(3.65 \pm .05) \times 10^5$ y	$(6.50 \pm .10) \times 10^{10}$ y
4	Specific Activity	$(3.79 \pm .05) \times 10^5$ y	
5	Relative to ^{240}Pu t_α	$(3.85 \pm .10) \times 10^5$ y	$(6.78 \pm .10) \times 10^{10}$ y
6	Relative to ^{239}Pu t_α	$(3.851 \pm .016) \times 10^5$ y	
7	Relative to ^{238}Pu t_α	$(3.675 \pm .07) \times 10^5$ y	
8	Calorimetric	$(3.763 \pm .009) \times 10^5$ y	
9	Relative to ^{238}Pu t_α		$(6.8 \pm .7) \times 10^{10}$ y
10	Specific Activity		$(7.45 \pm .17) \times 10^{10}$ y
This Exp	Relative to ^{239}Pu t_α	$(3.736 \pm .025) \times 10^5$ y	$(6.79 \pm .05) \times 10^{10}$ y
Unweighted Average		$(3.759 \pm .033) \times 10^5$ y	$(6.86 \pm .18) \times 10^{10}$ y
Weighted Average		$(3.776 \pm .007) \times 10^5$ y	$(6.78 \pm .04) \times 10^{10}$ y

[a]All measurements made relative to other Pu half-lives were adjusted to the appropriate half life in Table 2.

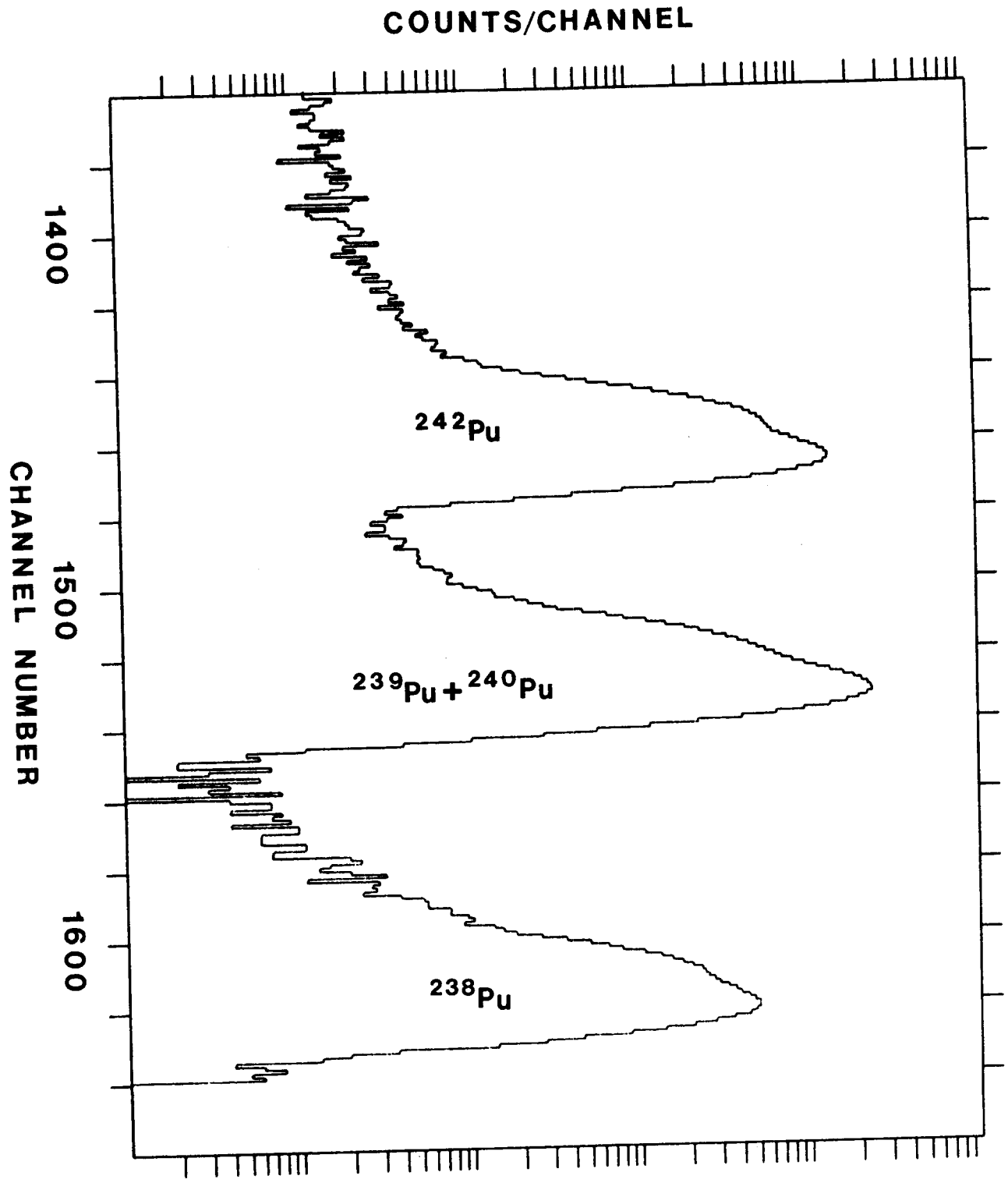

Fig. 1. The Alpha Particle Energy Spectrum for Sample 50. (ANL Neg. No. 111-77-909).

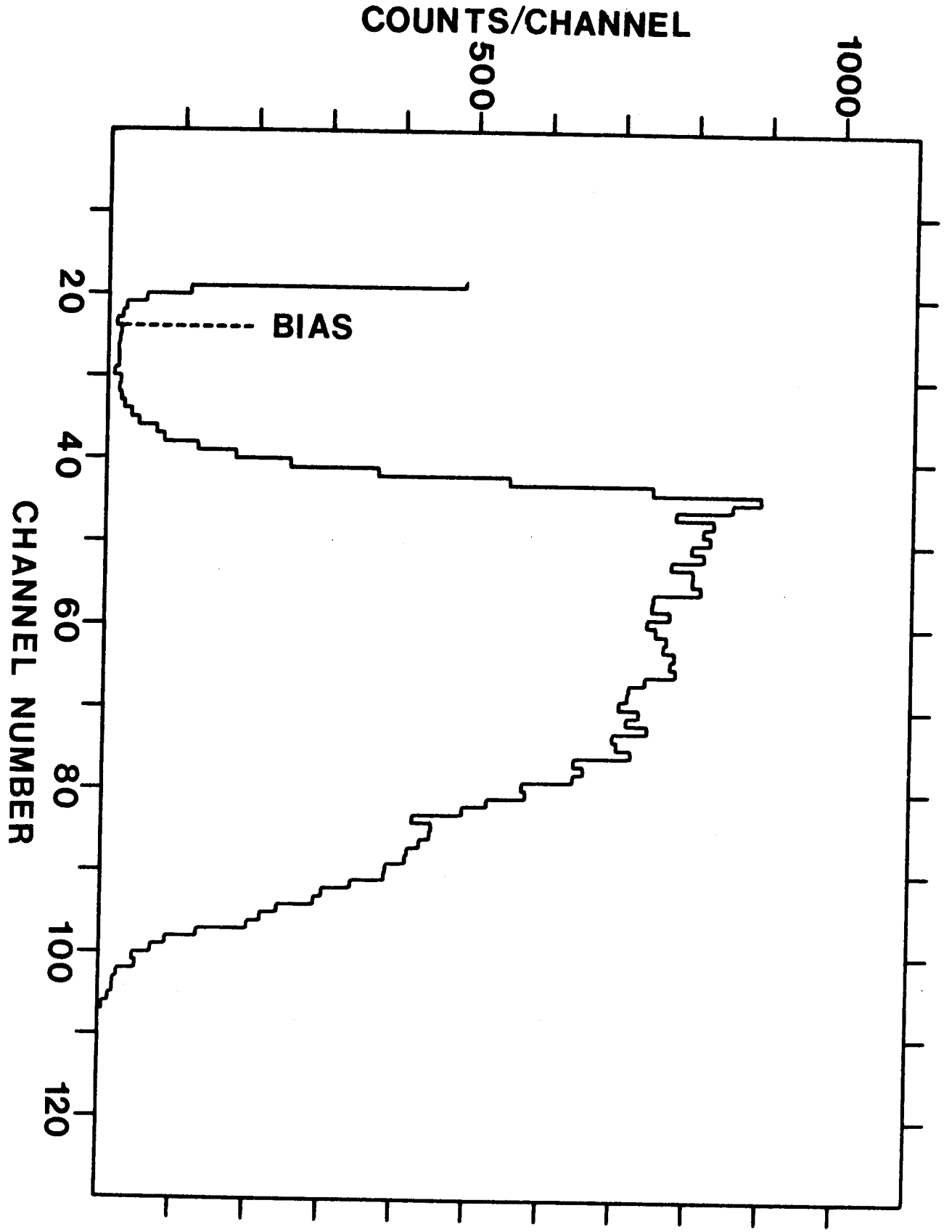

Fig. 2. The Fission Fragment Pulse-Height Spectrum for Sample 42. (ANL Neg. No. 116-77-920).