Spontaneous Fission Half-Life of Pu^{240}

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The spontaneous fission half-life of Pu^{240} has been measured, using ionization chamber and coincidence techniques, on sources containing microgram quantities of plutonium with 91.89 weight percent Pu^{240}. A new value of $(1.340\pm0.015)\times10^{11}$ years was obtained, which is higher than the previously accepted value.

1. INTRODUCTION

A NEW determination of the Pu^{240} spontaneous fission half-life was undertaken in support of a program on the measurement of the fission product yields.

Scrutiny of the published literature reveals that the absolute value of the half-life is still doubtful. Kinderman[1] measured the specific activity of samples containing 2.68 and 5.60% Pu^{240} by counting the alpha emission in a low-geometry counter and the fission particles in an ion chamber. Precautions were taken against absorption losses by using sources of less than 70 $\mu g/cm^2$ and a 0.55% correction for the spontaneous fission of Pu^{238} was made. Care was taken to insure that no alpha pulse pile-up occurred. The half-life was found to be $(1.314\pm0.026)\times10^{11}$ years.

Barclay *et al.*[2] obtained a result of $(1.62\pm0.04)\times10^6$ fission/g hr, corresponding to a half-life of 1.22×10^{11} years, from specific activity measurements on samples containing 3.86% and 5.58% Pu^{240}. In a less sensitive experiment, Chamberlain *et al.*[3] produced Pu^{240} by neutron irradiation of Pu^{239}. The very small percentages of Pu^{240} in the samples were deduced from known irradiation data, including an assumption of the value of alpha (the neutron capture to fission ratio). This gave a half-life of 1.2×10^{11} years, in apparently good agreement with Barclay's result, but since the assumed value of alpha was considerably higher than the currently accepted value the agreement between the two results must be coincidental. No corrections appear to have been applied for pile-up of alpha particles which must occur in view of the milligram quantities of plutonium on the sources. This could account for the shorter half-lives.

In the work presented here, corrections for alpha pile-up, absorption, and fissions from other plutonium isotopes are negligible.

2. PREPARATION AND ANALYSIS OF THE SOLUTION

The PuO_2 was dissolved in concentrated HNO_3 containing a trace of concentrated HF and converted to the chloride by repeated distillation with a constant boiling HCl solution. Am^{241} was removed by ion exchange,[4] the resultant Pu solution was weighed and aliquots taken for mass spectrometric analysis, alpha counting, and chemical determination of the plutonium content. Further diluted aliquots were used for fission counting.

Mass spectrometric analyses yielded the following isotopic abundance by weights: $6.50\pm0.05\%$ Pu^{239}, $91.89\pm0.05\%$ Pu^{240}, $1.54\pm0.03\%$ Pu^{241}, and $0.07\pm0.002\%$ Pu^{242}.

The alpha disintegration rate of an aliquot of the bulk plutonium solution was measured in a scintillation counter which was calibrated against a low-geometry proportional counter. The total plutonium content was calculated to be 703.5 ± 7 mg per ml from the mass spectrometric results, the disintegration rate, and the alpha half-lives of the plutonium isotopes.[4,5] A chemical determination of the plutonium in the solution was also made. This involved reduction with trivalent antimony and back titration with potassium iodate.[6] The result of 703.3 ± 2.0 mg Pu per ml is in excellent agreement with that obtained with the physical method. Three separate dilutions, each containing about 60 μg Pu^{240} per gram but known to a relative standard deviation of 1%, were prepared at one-month intervals for fission counting. The accuracy of the dilution factors was confirmed by counting aliquots of these dilutions in the calibrated alpha scintillation counter.

Two types of source were prepared for fission counting. Weighed amounts from 0.1 g to 1.0 g solution were deposited on one-inch diameter stainless steel disks and dried by infrared evaporation or by freeze-drying for 2π counting. Other sources in the same mass range were deposited on 40-$\mu g/cm^2$ VYNS plastic films coated with a conducting surface of 20-$\mu g/cm^2$ gold, or on 195-$\mu g/cm^2$ gold foil for 4π counting.

3. COUNTING

The fissions were counted by two methods.

(a) An ionization chamber with 2π geometry was filled to 20 cm Hg pressure with a mixture of argon plus 10% methane gas and operated on the fission

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plateau. The assembly is designed to accept a stainless steel disk source which is positioned under a hemispherical cathode of 5-cm radius.

(b) A 4π ionization chamber,[7] divided by the thin source into two 2π geometry chambers, was operated with argon-methane mixture at one atmosphere pressure. Pulses from each half and coincidence between these were recorded. The disintegration rate of the source N_T, is given by

$$N_T=(1/2\omega)\cdot(N_1N_2/N_{1,2}),$$

where N_1, N_2 are the counting rates of sides 1 and 2 and $N_{1,2}$ is the coincidence rate. ω, the solid angle subtended by each half of the counter, was verified to be within 1% of 2π by comparison with a standard alpha source. Hence the disintegration rate is given by $N_T=(N_1N_2/N_{1,2})$. This method partially corrects for possible self-absorption if the sources are not uniformly thin.

Both counters were surrounded by 8-in. thick boron-loaded paraffin wax to absorb neutrons from the cosmic-ray background but no observable change in the fission rate occurred.

4. ALPHA PILE-UP[8]

Precautions were taken to minimize alpha pile-up pulses which could contribute spurious counts to the fission rate. The pulse duration was reduced to 0.5 μsec by using electron collection and short amplifier time constants. The number of superimposed pulses required to give a count above the bias level was made as large as possible by choosing the chamber gas pressures such that the alpha particles spend only a small part of their range in the counter before entering the walls. This discriminates in favor of the fission fragments by increasing the ratio between pulse heights due to fissions and those due to alpha particles. Finally, the alpha activity of the source was kept to a minimum by using microgram quantities of sample, the limit being provision of an adequate fission rate. As a check on the success of these refinements a Pu^{239} source producing 8×10^6 counts/min of alpha activity was placed in the counter under normal fission counting conditions. No counts were observed in a two-hour run.

5. RESULTS AND CONCLUSIONS

The weighted mean of the results obtained for ten sources prepared by the variety of methods described in Sec. 2, yields a spontaneous fission half-life of $(1.340\pm0.008)\times10^{11}$ years. When this error is combined with the estimated uncertainty in the total plutonium determination, the value becomes

$$(1.340\pm0.015)\times10^{11} \text{ years.}$$

Self-absorption corrections for the 4π counter are quite negligible as the sources are only 2–20 $\mu g/cm^2$ thick. In the 4π counter some slight absorption occurs in the source backing, but the method of calculating the disintegration rate is self-correcting for this.

Since the fission rate calculated by the 4π method is dependent on the square of any possible alpha pile-up and the rate in the 2π counter is directly dependent on this, the agreement between the two methods indicates no spurious counts are occurring.

Fissions from Pu^{239} contribute a negligible correction but the Pu^{242} contributes 0.17% to the fission rate. This has been allowed for in the above result.

The half-life obtained in this work confirms Kinderman's value of $(1.314\pm0.026)\times10^{11}$ years.

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