Half-Lives of Pu^{240} and Pu^{239}†

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The half-lives of Pu^{240} and Pu^{239} have been determined by measuring the specific alpha activity and the Pu^{240} content of four plutonium samples which had received different amounts of neutron irradiation. With the assumption that Pu^{238} contributes 13 percent to the observed increase in alpha activity, the half-life of Pu^{240} is 6300±600 years. The half-life of Pu^{239} is found to be 24 400±500 years.

THE specific alpha activity and Pu^{240} content of four plutonium samples have been measured. All the plutonium samples were prepared from uranium which had been neutron-irradiated in a graphite lattice pile. The chemical procedure used to separate the plutonium from the uranium and fission products and to purify it is capable of removing all other elements from plutonium.

The specific alpha activity of the plutonium was determined as follows: A solution of pure Pu was assayed for alpha activity by mounting small aliquots (~0.25 μg Pu) on glass microscope cover slips and counting in a "50 percent geometry" ionization chamber connected to a linear amplifier. The solution was assayed for Pu by at least one of the following methods: (1) weighing PuO_2 made by igniting an aliquot of the solution; (2) titrating Pu^{III} to Pu^{IV} with ceric sulfate. The Pu^{III} was prepared by fuming an aliquot of the solution with H_2SO_4 and reducing the Pu with zinc amalgam[1]; (3) weighing a piece of pure plutonium metal before dissolving it to make the solution. Samples *A* and *B* were assayed by method (1). Samples *C* and *D* were assayed by all three methods. The three methods of assay gave very satisfactory checks.

The Pu^{240} content was determined by measuring the spontaneous fission rates of several ~0.5-mg samples of Pu, each mounted in an ionization chamber attached to a battery-operated linear amplifier. The Pu was mounted in a thin film on a platinum disk. The effective weight of Pu^{239} was determined by placing the chamber containing the Pu in a known slow neutron flux and measuring the neutron-induced fission rate. From previous calibrations against the mass spectrograph results the spontaneous fission rate was converted to weight of Pu^{240}.

TABLE I. Specific alpha activity of plutonium samples *vs* Pu^{240} content.

Sample No.	Specific alpha activity (counts/min per μg Pu)	Pu^{240} content (μg Pu^{240} per μg Pu)
A	69 480±100	0.00063±0.00006
B	69 960±100	0.0034 ±0.0002
C	72 510±100	0.0133 ±0.0006
D	72 930±100	0.0157 ±0.0006

The data are given in Table I and plotted in Fig. 1.

If all of the increase in the specific alpha activity is due to the presence of Pu^{240}, then the alpha activity of 0.01 μg of Pu^{240} is

$$[71\,650-(69\,240)(0.99)]\ \text{counts/min} = 3100\ \text{counts/min}.$$

Then if one assumes 51 percent geometry for the 50 percent geometry chamber, the specific alpha activity of Pu^{240} is 6.08×10^5 dis/min per μg Pu^{240} and the half-life of Pu^{240} is 5500 years.

However, according to Perlman and Seaborg,[2] 13 percent of the increase in specific alpha activity is due to the presence of Pu^{238}. This correction increases the result for the half-life of Pu^{240} to 6300 years. The estimated probable error for the half-life of Pu^{240} is 10 percent.

From the intercept of the specific alpha activity curve the half-life of Pu^{239} can be calculated. Assuming 51 percent geometry for the 50 percent geometry chamber, the specific alpha activity of Pu^{239} is 136 000

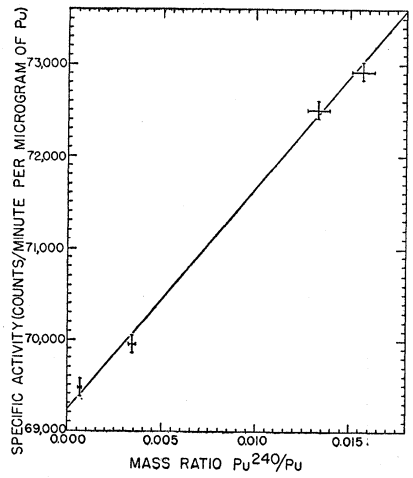

FIG. 1. Specific alpha activity of Pu^{240}—Pu^{239} mixtures *vs* mass ratio Pu^{240}/Pu for the mixtures.

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[1] We are indebted to Mr. Harold Boaz of group CM-9 for developing this method and performing the titration.

[2] I. Perlman and G. T. Seaborg, Los Alamos Report MUC-GTS-1872, July 28, 1945 (unpublished).

dis/min per μg of Pu^{239}, and the half-life of Pu^{239} is 24 400 years. The estimated probable error for the half-life of Pu^{239} is 2 percent, the uncertainty coming mainly from the uncertainty in the geometry.

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Evidence for Si^{32}, a Long-Lived Beta Emitter*,†

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Neutron-irradiated quartz has been found to contain 14.3-day P^{32} more than two years after the end of irradiation. This is evidence for a long-lived Si^{32} formed from stable Si^{30} by the capture of two neutrons. The ratio of half-life of Si^{32}, in years, to neutron capture cross section of Si^{31}, in barns is 600.

PREVIOUS work[1,2] has indicated that the unknown nuclide Si^{32} might be a long-lived beta emitter. This communication reports direct evidence (via isolation of its P^{32} daughter) for such a long lived Si^{32}.

Quartz, that had been intensively irradiated with thermal neutrons at the Hanford pile and allowed to cool for more than two years,[3] has been found to contain small amounts of 14.3-day P^{32}. The radiophosphorous was separated from approximately 100-gram samples of the quartz by volatilization of the silicon as SiF_4 from sulfuric and hydrofluoric acids in the presence of phosphate carrier. It was then purified by ammonium phosphomolybdate and magnesium ammonium phosphate precipitations. Included also were CuS scavengings and decontamination from vanadium by reduction of added V^{5+} carrier prior to some of the phosphate precipitations. The identification of the separated radioactivity as P^{32} was made from the decay and absorption characteristics of the radiations.

Table I lists the results on the phosphorous activity isolated from three samples of the quartz. The SiF_4 evaporation took up to two weeks. The carrier was added in several portions during the evaporation. Column three of the table lists the midpoint of this evaporation period. Column four gives the observed initial counting rate of the purified phosphorous. The chemistry extended over a period of 10 to 26 days and the chemical yields were between 15 and 50 percent. Finally, the last column lists the P^{32} activity of the original quartz calculated from the observed activity, the chemical yields, and the decay periods. As an additional check on radiochemical purity, the decay of the phosphorous sample IV was followed for 10 days and then subjected to an additional cycle of radiochemical purification. The result was no change (less than 5 percent) in specific activity and in the absorption characteristics.

TABLE I. P^{32} content of old neutron-irradiated quartz.

Sample	Wt of quartz used (g)	Date of SiF_4 evaporation	P^{32} activity observed (counts/min)	Specific activity of the quartz (counts/min g)
II	110	2/6/53	6.4	0.5
III	120	3/2/53	6.9	0.5
IV	120	5/6/53	31.0	0.8
			Weighted average	0.66

Several samples of unirradiated quartz, treated chemically in an identical manner, showed no P^{32} activity. Our limit of detection was about one-fiftieth of that observed in the most active sample isolated from the irradiated quartz.

Table I shows that there is P^{32} activity associated with neutron-irradiated quartz long after that formed directly by pile radiations has died away. In addition this activity is essentially constant over a period of about three months. A reasonable interpretation is that a long-lived parent, Si^{32}, has been formed from stable Si^{30} by the capture of two neutrons. From the weighted average of the specific activities indicated in Table I (0.66), the detection efficiency of our end-window proportional counters for P^{32} radiations ($\sim$45 percent), the irradiation conditions (flux and time), the thermal neutron capture cross section of Si^{30} (0.2 barn) and the half-life of Si^{31} (156 min), we calculate the ratio of half-life of Si^{32}, in years, to neutron capture cross section of Si^{31}, in barns, to be 600. The thermal neutron absorption cross section of Si^{31} is not known. For a cross section of 0.1 barn, the half-life of Si^{32} is calculated to be 60 years.

* Since preparation of this manuscript, evidence for the production of Si^{32} in the 340-Mev proton spallation of chlorine, has been presented by M. Lindner, Phys. **91**, 642 (1953).

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[1] M. Lindner, Phys. Rev. **89**, 1150 (1953).

[2] A. Turkevich and Althea Tompkins, Phys. Rev. **90**, 247 (1953).

[3] We thank Dr. F. T. Hagemann of the Argonne National Laboratory for making this quartz available to us.