Half-Lives and Neutron Capture Cross Sections of the Heavy Plutonium Isotopes

J. P. BUTLER AND T. A. EASTWOOD, *Atomic Energy of Canada, Ltd., Chalk River, Ontario, Canada*

AND

T. L. COLLINS, M. E. JONES, F. M. ROURKE, AND R. P. SCHUMAN, *Knolls Atomic Power Laboratory, Schenectady, New York**

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Specific alpha activities have been obtained for Pu^{240} and Pu^{242} corresponding to half-lives of 6600 ± 100 years and $379\,000\pm5000$ years, respectively. A half-life of $(7.5\pm2)\times10^7$ years has been obtained for Pu^{244} by separating uranium daughters. The irradiation of a sample of Pu containing Pu^{244} gave a neutron capture cross section of 2.1 ± 0.3 barns for Pu^{244} and yeilded 10.6 ± 0.4 hr Pu^{245} and its 124 ± 1 min Am^{245} daughter.

INTRODUCTION

AFTER Pu^{239} has been irradiated for a long time in a high-flux reactor, the resulting plutonium will contain all the long-lived plutonium isotopes from Pu^{238} to Pu^{244}, as well as other actinide nuclides up to Fm^{255}.[1,2] A long neutron irradiation of plutonium is an excellent way to produce material which contains mainly Pu^{242} and also detectable amounts of Pu^{244}. Small amounts of Pu^{242} and Pu^{244} have also been produced by the neutron irradiation of Am^{241} and Am^{243}, respectively.[3,4]

We have several samples of plutonium that have been irradiated for a very long time in the Materials Testing Reactor. One of them contains mainly Pu^{242} and an appreciable concentration of Pu^{244}. In order to confirm and extend data on the heavy isotopes of plutonium, determinations were made of the half-lives of Pu^{240} and Pu^{242} by specific alpha activity measurements, and of the capture cross section and half-life of Pu^{244}. Determinations were also made of the half-lives of Pu^{245} and Am^{245}.

SPECIFIC ALPHA ACTIVITIES

The plutonium samples were irradiated as plutonium-aluminum alloy. After irradiation, the plutonium and the actinides were separated and purified by methods described in a previous paper.[5] The plutonium was further purified by a series of Dowex 1 anion column separations and plutonium fluoride and plutonium peroxide precipitations. The final purified plutonium samples were isotopically analyzed using a surface ionization source and both a single- and a double-focusing mass spectrometer.[6] The results of the analyses on two samples, *B* and *C*, are given in Table I.

A sample of plutonium from *B* and one from *C* were dissolved in dilute HNO_3, then reduced with hydroxylamine hydrochloride to Pu^{+3}. The plutonium was then determined colorimetrically by Mr. A. J. Frey of Atomic Energy of Canada, Ltd., who compared the absorption of the Pu^{+3} in the unknown solutions with standard solutions of Pu. The method has an accuracy of better than $\pm1\%$.

After the plutonium concentrations were determined, known aliquots were taken, mounted on platinum counting disks, and alpha counted with proportional counters of known geometry. The counts were then corrected for backscattering, geometry, and coincidence loss, and the specific alpha disintegration rates of the plutonium samples determined.

Small samples of the plutonium were mounted for alpha pulse analysis by vacuum sublimation of the plutonium from a tantalum filament onto a polished platinum disk. The alpha groups were analyzed using the AECL gridded ionization chamber and a 30-channel pulse-height analyzer.[7] The spectra were spread out and analyzed in sections so that the valleys between peaks were well defined and corrections could be made for the small contributions due to scattering of the more energetic alpha groups. Figures 1 and 2 are typical alpha spectra obtained in this manner. From the alpha spectra, the percentage abundances of the 5.48-Mev

TABLE I. Isotopic composition and alpha abundances of MTR-irradiated plutonium.

	Sample *B*		Sample *C*	
Nuclide	Isotopic abundances	Alpha abundances	Isotopic abundances	Alpha abundances
Pu^{238}	$0.86\pm0.03\%$	69.97%	$0.076\pm0.005\%$	69.46%
Pu^{239}	$1.74\pm0.06\%$		$0.056\pm0.004\%$	
		$28.72\pm0.18\%$		$8.60\pm0.17\%$
Pu^{240}	$26.15\pm0.15\%$		$0.673\pm0.008\%$	
Pu^{241}	$15.4\ \pm0.3\%$		$0.217\pm0.008\%$	
		$1.31\pm0.06\%$		$21.94\pm0.10\%$
Pu^{242}	$55.9\ \pm0.4\%$		$98.90\ \pm0.02\%$	
Pu^{244}	$0.01\pm0.005\%$	...	$0.076\pm0.005\%$	...

* The Knolls Atomic Power Laboratory, Schenectady, New York, is operated for the U. S. Atomic Energy Commission by the General Electric Company.

[1] Studier, Fields, Sellers, Friedman, Stevens, Mech, Diamond, Sedlet, and Huizenga, Phys. Rev. **93**, 1433 (1954).

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alphas due to Pu^{238}, the 5.15-Mev alphas due to Pu^{240} and Pu^{239}, and the 4.88-Mev alphas due to Pu^{242} and Pu^{241} were obtained, see Table I. The contribution of Pu^{239} to the 5.15-Mev alpha group was calculated by using a Pu^{239} half-life of 24 400 years,[8] and the contribution of Pu^{241} to the 4.88-Mev alpha group was determined by using a Pu^{241} alpha half-life of 4×10^5 years.[3]

The specific activity of the 5.15-Mev alphas in sample *B*, when corrected for the small Pu^{239} contribution, corresponds to a half-life of 6560 ± 100 years for Pu^{240}. The half-life determined from sample *C* is 6710 ± 200 years. The probable errors include the errors in colorimetric analysis, aliquoting, counting and geometry determinations, alpha analysis, and mass analysis. Taking a weighted average of the two values gives a half-life of 6600 ± 100 years for Pu^{240}. This value is in excellent agreement with the better value of 6580 ± 40 years obtained by Inghram *et al.*[9] by mass

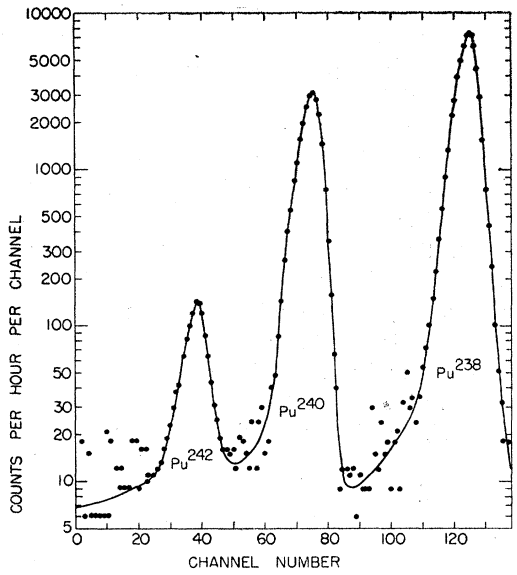

FIG. 1. Alpha energy spectrum of Pu sample *B*.

analysis of plutonium containing Pu^{239} and Pu^{240} and of the U^{235} and U^{236} daughters. Our value can also be compared with half-lives of 6300 ± 600 years,[10] 6240 ±120 years,[11] and 6760 years,[11] obtained by specific activity measurements.

The specific activity of Pu^{242} can be most accurately determined from sample *C* where the material is nearly pure Pu^{242}, the 4.88-Mev alpha abundance is high, and the correction due to Pu^{241} is insignificant. The half-life

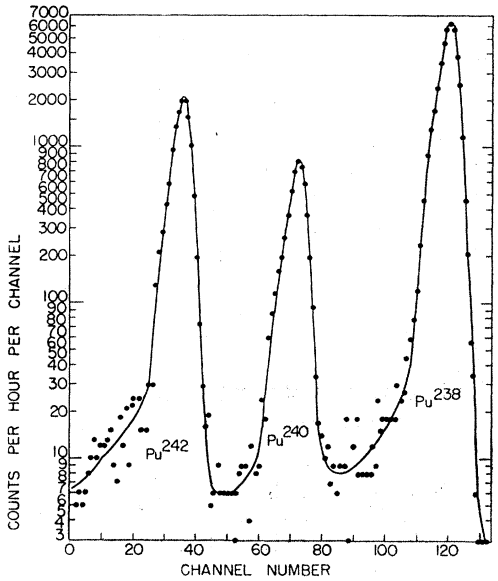

FIG. 2. Alpha energy spectrum of Pu sample *C*.

of Pu^{242} determined from sample *C* is $(3.79\pm0.05)\times10^5$ years. The error in the half-life is due to the colorimetric analysis, aliquoting, absolute alpha-counting, and alpha pulse analysis. The value is in excellent agreement with the value of $(3.73\pm0.05)\times10^5$ years obtained at Chalk River[12] by comparing the alpha and isotopic abundances of Pu^{242} and Pu^{238} in plutonium produced by the neutron irradiation of Am^{241}. Our value can also be compared with the value of $(3.9\pm0.1)\times10^5$ years[13] recently obtained by workers at the Argonne National Laboratory. The Pu^{242} half-life obtained from sample *B* is $(3.77\pm0.20)\times10^5$ years, assuming an alpha half-life of 4.0×10^5 years for Pu^{241}. Our results thus confirm the value of 4.0×10^5 years for the Pu^{241} alpha half-life.[3]

HALF-LIFE OF Pu^{244}

The plutonium *C* sample contains about 10 micrograms of Pu^{244}. This amount of Pu^{244} emits far too few alpha particles to be detected in the presence of the other plutonium alphas; however, the Pu^{244} decay rate can be determined by measuring the amount of U^{240} plus Np^{240} formed by its decay. The amount of U^{240} plus Np^{240} can be determined, in the presence of a much larger amount of U^{237} from Pu^{241} alpha decay, by separating uranium daughters and then following the decay of the uranium through sufficient absorber to cut out most of the weak U^{237} and U^{240} betas, but not many of the energetic Np^{240} betas. The half-lives and radiations of U^{240} and Np^{240} have been determined by Knight *et al.*[14] U^{240} has a 14.1-hour half-life and emits 0.36-Mev betas; Np^{240} has a 7.3-min half-life and emits

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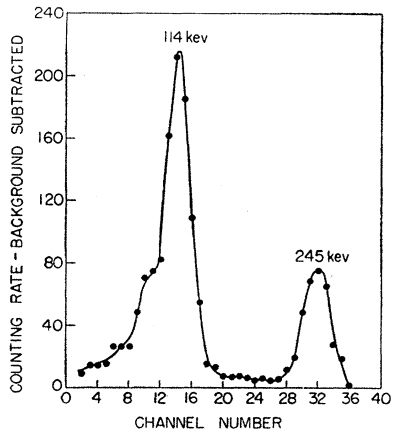

FIG. 3. Gamma spectrum of Am^{245}.

2.16 (52%), 1.59 (31%), 1.26 (11%), and 0.76 (6%) Mev betas and 0.56, 0.90, and 1.40 Mev gammas; 6.7-day U^{237} emits mainly 0.245-Mev betas and many weak gammas.[11]

For the determination of the Pu^{244} half-life, the plutonium was carefully purified and then uranium daughter activities were removed from the plutonium by absorption on a Dowex 1 column, see below. After about a day, uranium was again separated from the plutonium by passing a concentrated HCl solution of the Pu as Pu^{+3} through a Dowex 1 anion exchange resin column.[15] After the Pu had been removed from the column, the uranium was eluted with $\frac{1}{2}N$ HCl, the solution evaporated to a small volume, then taken up in concentrated HCl plus 1% HI and adsorbed on a second column. The final uranium was eluted from the second column and followed for decay through several different aluminum absorbers. Two components were found: a 6.7-day activity, due to U^{237} from Pu^{241} alpha decay, which was the predominant activity when counted without an absorber, and a 14-hour activity, due to Np^{240} in equilibrium with U^{240}, which formed more than half of the activity when counted through the thicker absorbers. Three uranium milkings were made and the decay was followed both on end-window G.M. counters and on an end-window proportional counter. From the amount of U^{240}, a half-life of $(7.5\pm2)\times10^7$ years was obtained for Pu^{244}. The values obtained from the different determinations agreed within $\pm7\%$; however, the errors in the counting efficiency determinations and the mass analysis increase the probable error to about $\pm25\%$. This half-life is in excellent agreement with the value of $(7.6\pm2)\times10^7$ years recently published by workers at the Argonne National Laboratory.[13] We have also obtained a rough check of the half-life by separating Np^{240} from the uranium and following its decay.

The amount of 6.7-day U^{237} milked from the plutonium has been determined roughly from the end window proportional counter decay curves. From the amount of U^{237}, an alpha half-life of $(3.8\pm0.6)\times10^5$ years can be calculated for Pu^{241}. The fact that this value agrees with the value obtained by alpha counting indicates that there was no large unmeasured loss of uranium during the milkings.

CROSS SECTION OF Pu^{244}—PROPERTIES OF Pu^{245} AND Am^{245}

Two nuclides, Pu^{245} and Am^{245}, have recently been reported in the literature by workers from Los Alamos[16] and the Argonne National Laboratory.[17] We have produced these nuclides by irradiating samples of Pu sample *C* in the NRX reactor. Two NRX pile irradiations were made, and a cobalt flux monitor included with one. After irradiation, the plutonium was purified by absorption and elution from a Dowex 1 anion-exchange resin column,[15] by LaF_3 precipitations, and finally by reabsorption as Pu^{+4} on a second Dowex 1 column. Americium was then milked at intervals from the plutonium absorbed on the column by elution with concentrated HCl plus ~1% HNO_3. The chemical yield of americium was found to be about 98%, and less than one part Pu in 10^5 was eluted. The amount of plutonium was determined after completion of the milkings by elution with concentrated HCl+HI and alpha counting.

After elution, the americium fractions were counted and followed for decay with either an end-window G.M. counter or an end-window proportional counter. Fifteen americium milkings were made on each irradiated sample, and the americium fractions were found to decay to background with an average half-life of 124±1 minutes. Feather analysis of aluminum absorption curves of the Am^{245} gave 0.88 Mev for the beta energy. A scintillation spectrogram (see Fig. 3) showed 0.114-Mev and 0.245-Mev gamma rays for Am^{245}. The energies are in agreement with those obtained by Los Alamos[16] and Argonne.[17] Our half-life of 124±1 min can be compared with 119±1 min obtained by Argonne[17] and 125±5 min obtained by Los Alamos.[16]

The amount of Pu^{245} and its half-life were determined from the amount of Am^{245} in the milked samples from the parent-daughter relationship,

$$A_1=\frac{\lambda_2-\lambda_1}{\lambda_2}A_2\frac{e^{-\lambda_1 t}}{e^{-\lambda_1 t}-e^{-\lambda_2 t}},$$

where A_1=activity of Pu^{245} at time of separation, A_2=activity of Am^{245} at time of separation, λ_1=decay constant of Pu^{245}, λ_2=decay constant of Am^{245}, and

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[16] Browne, Hoffman, Crane, Balagna, Higgins, Barnes, Hoff, Smith, Mize, and Bunker, J. Inorg. Nuclear Chem. **1**, 254 (1955).

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t= time between separation and previous separation. The decay constant determined for Pu^{245} corresponds to a half-life of 10.6±0.4 hours. The decay curve of Pu^{245}, determined from the Am^{245} milkings, showed a constant half-life for a period of more than ten half-lives. The Pu^{245} half-life can be compared with 10.5±0.5 hours obtained by Argonne[17] and 12±1 hours obtained by Los Alamos.[16]

The amount of Pu^{245} corresponds to an NRX pile capture cross-section of 2.1±0.3 barns for Pu^{244}. This value is a little higher than the value of 1.4±0.5 barns obtained by Argonne in the new CP-5 pile.[17]

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Excited States of S^{34}†*

G. S. STANFORD‡ AND G. F. PIEPER
Sloane Physics Laboratory, Yale University, New Haven, Connecticut
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The mode of decay of the second excited state of S^{34} has been examined, using a "slow-fast" coincidence technique to examine the gamma rays in coincidence with the appropriate proton group from the reaction $P^{31}(\alpha,p)S^{34}$. By comparing the observed cascade to cross-over intensity ratio with that calculated from the Weisskopf lifetime relationships, the number of possible spin-parity combinations for the first and second excited states can be restricted. When the results of beta decay studies are taken into account, it can be inferred that the first two excited states of S^{34} are both 2+. Spin 0+ for the ground state is assumed.

New measurements of the energies of the low-lying excited states of S^{34} are reported. It is concluded that there are levels at 2.13±0.02, 3.33±0.05, 4.32±0.08, and 4.8±0.1 Mev.

I. INTRODUCTION

PREVIOUS experiments[1–3] carried out in this laboratory have studied the decays of a number of excited states of Si^{30}, Mg^{26}, and Ne^{22}. In each case, the residual nucleus was obtained by means of an (α,p) reaction on the appropriate target. Each experiment consisted of selecting a particular excited state of the residual nucleus by selecting the proper proton group, and examining the energy spectrum of the gamma rays in coincidence with these protons. From such data it is possible to determine the mode of decay of an excited state: how much by cascade through other levels and how much by cross-over transitions. Such results are made meaningful by the theoretical lifetime relationships of Weisskopf,[4,5] which give the probability per unit time of a gamma-ray transition of given energy as a function of nuclear parameters and the multipole order of the radiation. The theoretical predictions are based on a single-particle model and are relatively crude, at times being in error by a factor of 100 or more; at the same time it can be pointed out that the relative transition probabilities can vary over a range of more than 10^{17}. Therefore, the relations are often useful in determining gamma-ray multipolarities.

The present paper reports the result of an investigation similar to the earlier ones concerning the second excited state of S^{34}, reached by means of the reaction $P^{31}(\alpha,p)S^{34}$. Mention is also made of those experimental techniques which have been improved since the earlier work.

II. EXPERIMENTAL METHODS

A. Target Preparation

The phosphorus targets were prepared by evaporating a solution of red phosphorus in ethyl alcohol, allowing the solute to deposit on a 0.0001-in. gold target backing at the bottom of a well one inch deep. The wall of the well was made of Teflon, which is not wet by alcohol, resulting in an even deposition of phosphorus on the gold.

The aluminum targets used for calibration consisted of aluminum foil of thickness ~0.2 mg/cm², fastened to a wire ring by means of indium solder.

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‡ Present address: Perkin-Elmer Corporation, Norwalk, Connecticut.

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