Plutonium-244 from Pile-Irradiated Plutonium

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THE high neutron flux of the Materials Testing Reactor (MTR) enhances by manyfold the possibility of multiple-order neutron-capture reactions. For plutonium-239 irradiations, Studier and Manning[1] in a detailed calculation have plotted the expected yields of mass numbers up to 244 as a function of integrated flux. In a program designed to follow up these calculations, several plutonium-239 samples have been irradiated in the MTR for progressively longer periods of time, the sample described here having an integrated flux of 4×10^{21} neutrons. Details of the experimental assembly in which the plutonium is irradiated are given elsewhere.[2]

The chemical procedure used was one which separated the plutonium from the *trans*-plutonium elements and the bulk of the fission products in the early stages of purification by utilizing the multivalency of plutonium. The separated plutonium fraction was then further purified by precipitations and solvent extractions.

The plutonium was analyzed in a 12-inch, 60-degree mass spectrometer using a multiple-filament source. The isotopic distribution agreed very well with the calculations of Fields and Weiss[3] in which they used more recent cross-section values than the earlier values used by Studier and Manning. In addition to the plutonium isotopes 239, 240, 241, 242, previously produced and identified in pile irradiations, this plutonium sample also contained Pu^{244}.[4] The Pu^{244}/Pu^{242} mole ratio was 0.0036 percent. Plutonium-244 is produced from Pu^{239} in the pile by the reactions shown in Fig. 1. The solid arrows represent the predominant reactions causing the production of the higher masses in the MTR. The dashed arrows indicate reaction paths of secondary importance. Plutonium-244 is formed by $Pu^{243}(n,\gamma)Pu^{244}$ reaction and possibly by electron capture of Am^{244}.

On the basis of a predicted alpha disintegration energy of 4.7 Mev for Pu^{244}, a closed cycle shows Pu^{244} to be approximately 1.3 Mev heavier than Cm^{244}. If the β^- energy of Am^{244} is greater than 1.3 Mev, it will be electron-capture unstable. If the β^- energy of Am^{244} is less than 1.3 Mev, Pu^{244} will be β^- unstable. In any case arguments from heavy element systematics indicate that the energies will be small (i.e., either the β^- energy of Pu^{244} or the electron-capture energy of Am^{244}). Other experiments have shown the β^- half-life of Pu^{244} to be greater than 1000 years[5] (the alpha half-life of Pu^{244} is estimated to be approximately 10^7 years). The capture cross section of Pu^{242} calculated from the data of this irradiation is 30 ± 10 barns in agreement with the earlier value.[6] Assuming all of the Pu^{244} to be produced by the $Pu^{243}(n,\gamma)Pu^{244}$ reaction and Pu^{242} to have a 30-barn capture cross section, the pile neutron-capture cross section of Pu^{243} is about 100 barns. On the other hand, if one assumes all the Pu^{244} to come from electron capture of Am^{244}, the electron-capture branching is approximately 0.5 percent. Assuming the tentative assignment[7] of a 25-minute β^- half-life for Am^{244} to be correct, the electron-capture half-life is greater than 3.4 days. Since a much longer electron-capture half-life would be predicted, it is reasonable to assume that most of the Pu^{244} comes from neutron capture by Pu^{243}. The possibility of energetic isomers of Am^{244} does, however, cast some doubt on the above assumption.

The technical assistance of C. H. Youngquist in the engineering aspects of these experiments is gratefully acknowledged. We also wish to thank W. M. Manning for many stimulating discussions.

[1] M. H. Studier and W. M. Manning (unpublished).
[2] A. B. Shuck (unpublished).
[3] P. R. Fields and M. A. Weiss (unpublished).
[4] This isotope was previously discovered by Hess, Fried, Pyle, and Inghram (unpublished).
[5] Fried, Pyle, and Fields (private communication).
[6] Pyle, Fields, and Huizenga (unpublished).
[7] Street, Ghiorso, and Seaborg, Phys. Rev. **79**, 530 (1950).

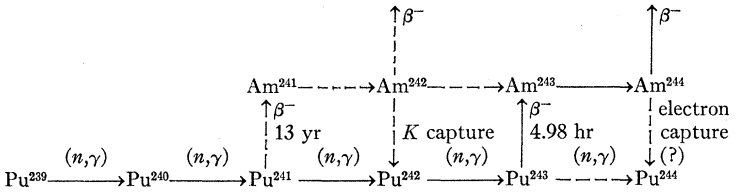

FIG. 1. Reactions in pile-irradiated Pu^{239}.

A New Isomer in Lead

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IN irradiations of Tl with 26-Mev deuterons, a new 3.5 ± 0.1 hr activity appeared in the Pb fraction. Its excitation curve, compared with those of 1.1-hr Pb^{204*} and 2.3-day Pb^{203} pointed to the reaction $Tl^{203}(d,3n)Pb^{202*}$. This activity has been studied with NaI scintillation spectrometers and a β-ray spectrometer. The γ rays, found in this isomer, are collected in Table I, together with their assignments, [based on $K/(L+M)$ ratios, conversion coefficients and half-lives] and the relative intensities of the transitions (γ rays+conversion electrons).

A decay scheme, derived from these data, is shown in Fig. 1. This decay scheme is consistent with the results of γ-γ coincidence measurements made with two scintillstion spectrometers in coincidence. The relative position of the 956-kev and 416-kev transitions is not determined; comparison with the first excited state in Pb^{204} and Pb^{206} suggests that the 416-kev transition should be the lower one. An $E1$ γ ray of 200 kev with an intensity of about 8 percent should also be present. Its K conversion line coincides

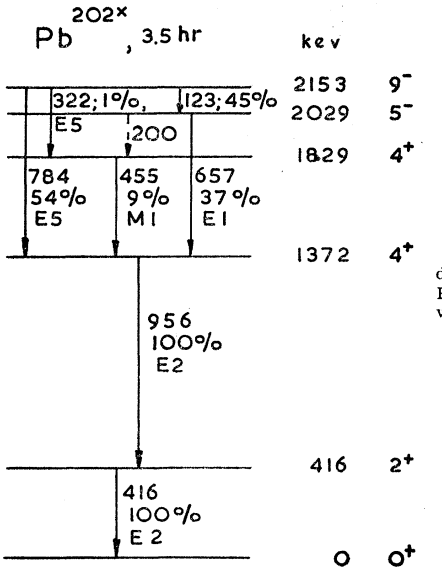

FIG. 1. Proposed decay scheme of Pb^{202*} (energy values in kev).

with the strong M conversion line of the 123-kev γ ray, and the L conversion line with the 196-kev conversion line of Pb^{203}, which was also present in the sample; in the scintillation spectrum the 277-kev Pb^{203} γ ray and the backscattering peak hinder the detection of this γ ray.

A more extensive report will be published in Physica. We thank Dr. C. J. Bakker and Dr. A. H. W. Aten, Jr., for their interest,

TABLE I. γ rays in Pb^{202*}.

Energy (kev)	Assignment	Intensity
x_K	—	~15
123±2	$E3,4$	45
322±4	$E5$	1.2
416±3	$E2$	102
455±4	$M1(+E2?)$	9
657±5	$E1$	40
784±4	$E5$	54
957±5	$E2$	98

R. H. Nussbaum, G. J. Nijgh, and L. T. M. Ornstein for help with the measurements, Mrs. T. de Vries-Hamerling for performing the chemical separations, and the Foundation for Fundamental Research of Matter (F.O.M.) of the Organization for Pure Scientific Research (Z.W.O.) for their support.

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Errata

Interpretation of Electron Scattering Experiments, L. I. SCHIFF [Phys. Rev. 92, 988 (1953)]. The last sentence at the end of the first paragraph: "This effect was noted earlier by M. Goldhaber and A. W. Sunyar [Phys. Rev. 83, 906 (1951)]" should have been placed at the end of footnote 3B.

Neutron Capture γ Rays from Scandium, Vanadium, Manganese, Cobalt, and Copper, G. A. BARTHOLOMEW AND B. B. KINSEY [Phys. Rev. 89, 386 (1953)]. Because of a regrettable oversight, the intensities for the vanadium γ rays given in Tables II and III of this paper are too large by a factor of 1.56. However, this error does not apply to Fig. 4. We are indebted to Dr. P. S. Mittelman for drawing our attention to this point.

Effects of the Atmosphere on the Penetrating Cosmic Radiation, ROBERT L. CHASSON [Phys. Rev. 89, 1255 (1953)]. In Table I, the signs of the temperature coefficients of Duperier and Chasson should be + rather than −.

The Energy Loss of a Fast Charged Particle by Čerenkov Radiation, R. M. STERNHEIMER [Phys. Rev. 91, 256 (1953)]. In this paper a dimensionless quantity b_p was defined as $b/(c/\nu_p)$, where b is the impact parameter. This definition should be $b/(c/2\pi\nu_p)$. All of the equations are unaffected, in particular Eqs. (1), (35), and (36) for the Čerenkov loss W_b. However, Eq. (24) for κ_p gives the absorption coefficient for a length $c/2\pi\nu_p$ instead of c/ν_p. The numerical values of b_p for the examples considered are increased by a factor 2π. As a result W_b is smaller than the values given in the paper. The case of emulsion [also reported in Phys. Rev. 89, 1148 (1953)] was recalculated using $b_p=31.4$. This gives $W_b(\infty)\cong 0.4\times 10^{-3}$ Mev/g cm^{-2}. The values of W_b for gases given in Table II should be decreased by $\frac{2}{3}Af_i \ln(2\pi)$, which is 0.104 Mev/g cm^{-2} for H_2, 0.052 Mev/g cm^{-2} for He, and 0.0135 Mev/g cm^{-2} for O_2 (model II). The resulting values of $W_b(\infty)$ (in Mev/g cm^{-2}) are 0.128 for H_2, 0.088 for He, and 0.0165 for O_2. $W_b(\infty)$ for Xe becomes 0.058 Mev/g cm^{-2}. Figure 1 for the Čerenkov loss J in emulsion pertains to $b=0.02\mu$ (instead of 0.13μ) and Fig. 3 pertains to $b=0.013$ cm (instead of 0.081 cm).

In the second line below Eq. (6), ν_{18} should be ν_{14}.

A Precision Measurement at 24 500 Volts of the Conversion Constant λv, GAELEN L. FELT, JOHN N. HARRIS, AND JESSE W. M. DUMOND [Phys. Rev. 92, 1160 (1953)]. The title should read: "A Precision Measurement at 24 500 Volts of the Conversion Constant λV."

The Scattering of Fast Neutrons by Iron, Lead, and Chromium, M. A. ROTHMAN, D. W. KENT, AND C. E. MANDEVILLE [Phys. Rev. 92, 1097 (1953)]. The word "unresolved" on the next to the last line of Abstract P6 should read "resolved."

Effect of Traps on Carrier Injection in Semiconductors, H. Y. FAN [Phys. Rev. 92, 1424 (1953)]. The factor τ_g appearing in the last section on the drift of injected carriers should be τ_f. The term dn_t/dt in Eqs. (18) and (26) should be replaced by $(R_{vt}-R_{tv})$, if the electron transitions between the traps and the conduction band were to be taken into account. In that case, the coefficient of Δn_t in (21) and the coefficient of Δp in (27) will become $[(1/\tau_f\tau_r)+(1/\tau_c\tau_t)]$ instead of $(1/\tau_f\tau_r)$, where $1/\tau_c=r_c(n_0+n_1)$.

A Binding Energy Calculation on He^4 with Single-Particle Wave Functions, P. G. WAKELY [Phys. Rev. 90, 724 (1953)]. The third square bracket in the wave function Ψ_2 should read $[\sqrt{\frac{1}{2}}\Phi(sp[2]^{13}P,sp[2]^{13}P,^{11}S)-\sqrt{\frac{1}{2}}\Phi(sp[2]^{31}P,sp[2]^{31}P,^{11}S)]$. The state referred to earlier as $(9s)^2(2p)^2[4]^{11}S$ should of course be $(1s)^2(2p)^2[4]^{11}S$.

Multiple Production of Pions in Nucleon-Nucleon Collisions at Cosmotron Energies, E. FERMI [Phys. Rev. 92, 452 (1953)]. In computing the statistical weights of the various states discussed in this paper, a factor $1/n!$ (n = number of pions) was omitted. For this reason, the statistical weights given in column 2 of Table II and in columns 2 and 3 of Table III should be divided by the factorials of the number of pions given in column 3 of Table II and column 4 of Table III. Corresponding changes should be made in the computed probabilities for the two cases. This correction has the effect of reducing the probability of events with high multiplicity. For example, for a neutron-proton collision, the probabilities of stars with 1, 3,