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.

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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).

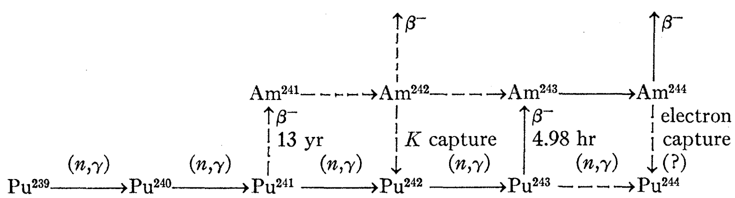

FIG. 1. Reactions in pile-irradiated Pu^{239}.

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