NEW SHORT-LIVED AMERICIUM BETA EMITTERS*

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We have chemically separated ²⁴⁷Am from the products of α -particle bombardment of ²⁴⁴Pu. Gamma transitions of 226 and 285 keV are associated with its decay. The intensities of the K and L conversion electrons from the 226-keV transition are most consistent with M2 multipolarity. We have found evidence in the americium fraction from both ³He- and α -particle bombardments of ²⁴⁴Pu for 153-, 205, and possibly 99- and 555-keV γ rays and believe that these transitions follow β decay of a high-spin ²⁴⁶Am isomer.

The neutron-rich americium isotopes have been identified only through mass 246, but consideration of nuclear-energy systematics and the appropriate Nilsson orbital assignments indicates that ²⁴⁷Am should have a half-life in the range of 10 to 60 min. Furthermore, in addition to the presently known 25-min isomer (I=2), ²⁴⁶Am may be expected to have low-energy high-spin states (I=7) with the configurations $(pn, \frac{5}{2} [523] \frac{9}{2} [734])$ or $(pn, \frac{5}{2} + [642] \frac{9}{2} [734])$, one of which may exist as an isomeric state. If this hypothetical isomeric state were to decay solely by β^- emission, a half-life in the range of 15 to 180 min can be estimated from the considerations mentioned above. These isotopes, together with 2.07-h ²⁴⁵Am, could be produced by 28-MeV α -particle bombardment of ²⁴⁴Pu by (α, p) , (α, pn) , and $(\alpha, p2n)$ reactions, although the cross sections might be somewhat less than 1.5 to 5 mb which was found by Vandenbosch et al.¹ for similar reactions in ²³⁵U and ²³⁸U. ³He bombardment of ²⁴⁴Pu should produce ²⁴⁶Am ²⁴⁵Am, and ²⁴⁴Am, but not ²⁴⁷Am. Sufficient ²⁴⁴Pu for such experiments was not available until recently, when 1.5 mg of a plutonium sample containing 74.2 at.% ²⁴⁴Pu, 25.3 at.% ²⁴²Pu, and 0.5 at.% of masses 238 through 241 was made available to us through the transuranium production program of the Division of Research of the U.S. Atomic Energy Commission. The material was produced by neutron irradiation of ²⁴²Pu in the High Flux Isotope Reactor followed by Calutron separation at Oak Ridge National Laboratory to further enrich the plutonium in mass 244.

In this note we report evidence for a new isotope, ²⁴⁷Am, made by 28-MeV α -particle bombardment of ²⁴⁴Pu. In addition, we have obtained evidence for a previously unreported 40-min americium activity which we tentatively assign as a high-spin isomer of ²⁴⁶Am.

Plutonium targets, prepared by evaporating 100 to 200 μ g of the plutonium on 4-mil alu-

minum plates, were bombarded with $\approx 2 \ \mu$ A-h of 28-MeV α particles in the Los Alamos variable-energy cyclotron. A similar bombardment with $\approx 4 \ \mu$ A-h of 25-MeV ³He particles was also performed. The plutonium was evaporated on 1-mil platinum for bombardment with $\approx 50 \ \mu$ A-h of 28-MeV α particles.

After bombardment, the major portion of the plutonium and reaction products was dissolved away from the target backing; the americium was carried on La(OH), precipitated first with NaOH and then with NH₄OH. After further purification by an oxidation procedure based on that described by Moore,² the americium was reduced to the trivalent state and passed through an anion resin column in concentrated HCl solution. The effluent liquid, ≈ 1 ml, was used directly as a source. Alternatively, after the initial hydroxide precipitations, the americium was purified by elution from a cation exchange resin column with a 20% ethanol-HCl mixture.³ Again, the solution was passed through an anion column and in the case of the 50- μ A-h bombardment, sulfide scavenge and LaF, precipitation steps were added. A sample for the observation of electron spectra was prepared by final evaporation of the produce solution on 0.5-mil Teflon film.

The γ -ray spectra were measured with a 7-mm×4-cm² Ge(Li) detector. Figures 1 and 2 show low-energy spectra of the americium activities produced by 28-MeV α -particle bombardment and by 25-MeV ³He-particle bombardment of the ²⁴⁴Pu. The half-lives of the 226and 285-keV γ rays shown in Fig. 1 were determined to be 24±3 min by following the decay of the individual photopeaks over a period of \approx 2 days. The K x rays also show an intense 24-min component. Although these half-lives are nearly the same as those of ²⁴⁴Am (I=1) and ²⁴⁶Am (I=2), the radiations reported⁴⁻⁶ for these nuclides include very few K x rays



FIG. 1. Low-energy γ -ray spectrum of americium nuclides produced by bombardment of ²⁴⁴Pu sample with 28-MeV α particles for 22 min. The spectrum was obtained with a 7-mm×4-cm² Ge(Li) detector in a 20-min count begun 39 min after the end of bombardment. The 252-keV γ ray and \approx 50% of the Cm K x rays are due to 2.07-h ²⁴⁵Am.

and no 226- or 285-keV γ rays. The 226- and 285-keV γ rays are absent from the spectrum of the americium activities produced in the ³He bombardment (Fig. 2). The 153- and 205keV γ rays shown in Fig. 1 decayed with a 40 \pm 7-min half-life, while those produced in the ³He bombardment (Fig. 2) decayed with an additional component due to the well-known 10-h ²⁴⁴Am isomer produced by the reaction ²⁴⁴Pu(³He, p2n). Spectra from both ³He- and α -particle bombardments show evidence for 99- and 555keV γ rays which decay with an \approx 40-min halflife. The γ -ray data are summarized in Table I.

Figure 3 shows an electron spectrum of the americium activities produced by the 28-MeV α bombardment of the ²⁴⁴Pu. The electron lines at 98 and 124 keV are assigned as the K shell internal conversion-electron lines from 226-and 252-keV γ transitions. Weaker lines at 202, 220, and 228 keV are probably the L-and M-shell conversion electrons from the 226- and 252-keV γ transitions. The prominence around channel 110 may be due to L-shell conversion of a 99-keV transition.

By comparison with electron and photopeak intensities of the ²⁴⁵Am 252-keV transition (α_K =1.9),⁷ α_K for the 226-keV transition was estimated to be 6±3. M2 multipolarity is indi-



FIG. 2. Low-energy γ -ray spectrum of americium nuclides produced by bombardment of ²⁴⁴Pu sample with 25-MeV ³He particles for 30 min. The spectrum was obtained with the Ge(Li) detector in a 10-min count begun 51 min after the end of bombardment. The 252keV γ ray and most of the Cm K x rays are due to 2.07h ²⁴⁵Am.

cated. The energy and probable M2 multipolarity of this 226-keV transition suggest that it is identical to the previously reported 226keV γ transition from the delayed level ($t_{1/2}$ = 24 μ sec)⁸ in ²⁴⁷Cm. If, as in ²⁴⁹Cf (151 neutrons), the $\frac{9}{2}$ [734] configuration⁹ describes the ²⁴⁷Cm ground state, then the $\frac{5}{2}^+$ [622] orbital best characterizes the 226-keV level. ²⁴⁷Am. either $\frac{5}{2}$ ⁺[642] or $\frac{5}{2}$ ⁻[523], should decay to this [622] band with $\log ft \approx 6.3$. If the 285-keV transition were M1, the conversion-electron peaks should have been comparable in intensity with those from the 226-keV transition (Fig. 3). The fact that they were not observed indicates that the transition is E1 or E2. The position of the 285-keV transition in the ²⁴⁷Cm level

Table I. Summary of γ -ray data.

$24 \pm 3 \min {}^{247}$ Am		$40 \pm 7 \min {}^{246} \mathrm{Am}$	
E (keV)	Relative γ-ray intensity	E (keV)	Relative γ-ray intensity
226 ± 2	0.25 ± 0.07	99 ± 2	0.2 ± 0.1
285 ± 2	1.00	153 ± 2	0.6 ± 0.2
		$\begin{array}{c} 205\pm2\\ 555\pm5\end{array}$	$1.00\\1.6\pm0.3$



FIG. 3. Electron spectrum of americium nuclides produced by bombardment of ²⁴⁴Pu sample with 28-MeV α particles for 32 min. The spectrum was measured with a cooled 2-mm×80-mm² Si(Li) detector in a 15min count begun 55 min after the end of bombardment. The prominence around channel 110 is probably from *L*-shell conversion of a 99-keV transition from ²⁴⁶Am.

structure cannot be ascertained from our present data.

The 153- and 205-keV γ rays associated with the 40-min americium activity have the same energies that we observe in the decay of 10h ²⁴⁴Am. The $8^+(502 \text{ keV})$ and $6^+(296 \text{ keV})$ members of the ²⁴⁴Cm ground-state rotational band are de-excited by 206- and 154-keV transitions,⁴ and the levels in ²⁴⁶Cm should be nearly identical. Either of the two possible I=7 isomeric states of ²⁴⁶Am should undergo β decay predominantly to the ²⁴⁶Cm quasiparticle state $(nn, \frac{9}{2}[734]\frac{7}{2}[624])_{8}$, calculated to be at 1100 keV.¹⁰ This state is expected to be de-excited by an E1 transition to the 8^+ member of the ²⁴⁶Cm ground-state band. Our source strength was too weak to permit $\gamma - \gamma$ coincidence measurements, but on the basis of γ -ray energy and intensity measurements (Table I) it appears that a 555-keV transition de-excites a ²⁴⁶Cm level at 1055 keV to the 8⁺ member of the groundstate band. There is also evidence for the 99keV transition expected to de-excite the 4⁺ member (142 keV). The conversion electrons from the highly converted 43-keV transition depopulating the 2⁺ states in ²⁴⁶Cm and ²⁴⁴Cm could

not be detected in our experimental arrangement.

In summary, our evidence for observation of ²⁴⁷Am and a high-spin isomer of ²⁴⁶Am is as follows:

(1) The new activities followed the americium fraction through alternative types of chemical purification procedures, and the energies of the observed K x rays are identical to those of curium, thus indicating that the activities are americium β emitters.

(2) The new 24-min americium activity has been assigned to mass 247 on the following bases: (a) It was produced in α -particle bombardment, but not in ³He bombardment of ²⁴⁴Pu. (b) The energy and M2 multipolarity of the 226keV transition are consistent with the energy and 24- μ sec half-life of the 226-keV state⁸ in ²⁴⁷Cm. (c) ²⁴⁷Am with Ω^{π} either $\frac{5}{2}^+$ or $\frac{5}{2}^-$ is expected to strongly β^- populate this 226-keV ($\frac{5}{2}^+$ [622]) band in ²⁴⁷Cm. (d) It cannot be due to 25-min ²⁴⁴Am or ²⁴⁶Am since 226- and 285-keV γ rays are not observed in their decay.⁴⁻⁶

(3) The 40-min americium activity has been tentatively assigned to a high-spin isomer of ²⁴⁶Am on the following bases: (a) The activity was produced by both ³He- and α -particle bombardments of the ²⁴⁴Pu. (b) The energies of the observed 153- and 205-keV γ rays are identical to the energies of the transitions expected to de-excite the 8⁺ and 6⁺ members of the ²⁴⁶Cm ground-state band. There is also evidence for a 99-keV transition which should depopulate the 4⁺ level of this band.

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¹R. Vandenbosch, T. D. Thomas, S. E. Vandenbosch, R. A. Glass, and G. T. Seaborg, Phys. Rev. <u>111</u>, 1358 (1958).

²F. L. Moore, Anal. Chem. <u>35</u>, 715 (1963).

³S. G. Thompson, B. G. Harvey, G. R. Choppin, and G. T. Seaborg, J. Am. Chem. Soc. <u>76</u>, 6229 (1954).

⁴S. E. Vandenbosch and P. Day, Nucl. Phys. <u>30</u>, 177 (1962).

 ${}^{5}\mathrm{F.}$ S. Stephens, F. Asaro, S. Fried, and I. Perlman, Phys. Rev. Letters <u>15</u>, 420 (1965).

⁶C. J. Orth, Phys. Rev. 148, 1226 (1966).

⁷M. E. Bunker, D. C. Hoffman, C. J. Orth, and J. W. Starner, Nucl. Phys. A97, 593 (1967).

 8 R. J. Silva, A. Chetham-Strode, Jr., J. R. Tarrant, and I. R. Williams, Bull. Am. Phys. Soc. <u>11</u>, 408 (1966).

⁹F. S. Stephens, F. Asaro, S. G. Thompson, and I. Perlman, Bull. Am. Phys. Soc. <u>2</u>, 394 (1957). ¹⁰V. G. Soloviev and T. Siklos, Nucl. Phys. <u>59</u>, 145 (1964).

ANGULAR DISTRIBUTIONS FOR THE INVERSE PHOTONUCLEAR PROCESS IN Si²⁸ IN THE EIGENCHANNEL REACTION THEORY*

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Using the eigenchannel reaction theory we performed coupled-channel calculations for Si^{28} and computed the differential cross section for $Al^{27}(p,\gamma_0)Si^{28}$ over the energy range 6 MeV < E_p < 16 MeV. The obtained angular distributions are nearly constant over the whole energy range and agree with the experiment in that they are almost isotropic. Thus, it seems that in this framework we can give a natural explanation for the peculiar behavior of the $Al^{27}(p,\gamma_0)Si^{28}$ cross section.

During the past four years the inverse photonuclear process in Si²⁸, and, in particular, the reaction $Al^{27}(p, \gamma_0)Si^{28}$ leading to the ground state of Si²⁸ has gained great interest because of its surprising feature that the gamma-ray angular distribution approximately remains constant throughout the region of the giant resonance, in spite of the fact that the yield curves show considerable structure, especially for high-energy resolution.¹⁻³ As a matter of fact, the Al²⁷ (p, γ_0) Si²⁸ angular distributions are almost isotropic through the whole energy range. This behavior distinguishes them clearly from γ -ray angular distributions observed for $B^{11}(p,$ γ)C^{12,12*4} or $F^{19}(p, \gamma)$ Ne^{20,20*,5} for example, showing a rather pronounced energy-dependent peaking.

It was demonstrated in Ref. 3 that the Al²⁷(p, γ_0)Si²⁸ 90° yield curve exhibits Ericson fluctuations which can be characterized by peaks of 50-keV widths which are spaced about 100 keV apart. Figure 8 of Ref. 3 shows that the integrated (p, γ_0) cross section smoothed by using an averaging interval of 600 keV has a remarkable intermediate structure with peaks at 18.2-, 19.0-, 19.8-, and 21.3-MeV excitation energy, respectively. These intermediate-resonances are expected to be predictable by a 1p-1h treatment of the compound nucleus. From the conventional shell-model calculation of Bolen and Eisenberg,⁶ however, one could obtain no indication for the constancy of the γ -ray angular distribution. Therefore, Singh et al.³ concluded that the 1⁻ nuclear eigenstates of \overline{Si}^{28} which produce the intermediate resonances should not be responsible for the angular distribution, but that it would result from some sort of single-proton configuration, characteristic for the giant resonance as a whole.

We do not agree with this view, but intend to show that the observed angular distribution can, indeed, be explained by a shell-model calculation in the 1p-1h Tamm-Dancoff approximation provided that the particle continua are treated carefully. The procedure we actually use for accurately solving the coupled-channelcontinuum problem is known as the eigenchannel method⁷ and was first worked out and successfully employed in the case of $O^{16}.^{8-10}$

The basic idea of the eigenchannel method is to construct a complete set of degenerate nuclear scattering states to a given excitation energy $E_{\rm exc}$ of the compound system; thus they are eigenfunctions of the nuclear 1p-1h Hamiltonian as well as of the S matrix. These eigenchannel functions contain real radial wave functions in the various channels. Therefore, boundary conditions on a distant surface can easily be formulated and a discretization of the particle continuum states can be obtained. Then, the nuclear Hamiltonian can be diagonalized similarly as for pure bound configura-