Half-life of the electron capture decaying isotope ^{236}Am

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The neutron deficient americium isotope ^{236}Am has been produced via the ^{235}U(^{6}Li, 5n) reaction. The reaction products were transported to an ion source of the on-line isotope separator (JAERI-ISOL) by a He/KCl aerosol jet transport system, and americium atoms were successfully ionized and mass-separated. PuK x rays associated with the EC decay of Am nuclei at the mass fraction of 236 were observed, indicating the definite identification of ^{236}Am. The half-life of ^{236}Am is determined to be 4.4 ± 0.8 min and the production cross section is estimated to be about 70 μb. [S0556-2813(98)03704-2]

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There still remain many isotopes to be discovered in the region of neutron deficient actinides. Decay properties of these nuclides lead to considerable advances in the understanding of proton excess heavy nuclei: verification of the proton drip line, nuclear structure of large deformed nuclei such as hexadecapole deformation, and fission barrier heights of neutron deficient nuclei far from stability. Electron-capture delayed fission (ECDF) which is an exotic nuclear decay process is also expected to occur in the neutron deficient actinide region, and the probability of ECDF was reported in some odd-odd neptunium, americium, and berkelium nuclei [1–4].

The aim of the present study is to determine the half-life of the neutron deficient americium isotope ^{236}Am which would decay dominantly through EC. Although Hall *et al.* tried to identify ^{236}Am by the radiochemical method [5], there remains some ambiguity on the determination of mass number.

We have developed a composite system consisting of a gas-jet transport apparatus and a thermal ion source in the on-line isotope separator (JAERI-ISOL) [6]. This gas-jet coupled JAERI-ISOL system enables us to determine simultaneously mass number via the isotope separator and atomic number by the measurements of x rays associated with the EC/$\beta^{\pm}$ decay of a nucleus. Some new neutron-rich rare-earth isotopes produced in the proton-induced fission of ^{238}U were identified with this system [7,8]. In this Report, the half-life of ^{236}Am produced in the ^{235}U(^{6}Li,5n) reaction is measured with this system and the decay properties of ^{236}Am will be briefly discussed.

Experiments were performed at the JAERI (Japan Atomic Energy Research Institute) tandem accelerator facility. A stack of fifteen ^{235}U (93% enrichment) targets set in the multiple-target chamber was bombarded with the ^{6}Li^{3+} beams with the intensity of 100–300 particle nA. Each target was prepared by electrodeposition onto a 0.8 mg/cm^2 thick aluminum foil, and the thickness estimated by α spectrometry was about 200 μg/cm^2. The performance of the present system including the gas-jet transportation and the ionization of Am atoms was examined by using the isotope ^{237}Am produced in the ^{235}U(^{6}Li,4n) reaction with an energy of 39–43 MeV. ^{236}Am was produced via the ^{235}U(^{6}Li,5n) reaction at 46–50 MeV.

The reaction products recoiling out of the targets were stopped in He gas (71 kPa) loaded with KCl clusters produced by sublimation from the surface of KCl powder at 650 °C. The products attached to the aerosols were swept out of the target chamber with the He gas (0.8 l/min) into a capillary (1.4 mm in diameter and 8 m length). The transported nuclides were ionized in the thermal ion source at 2450 K [6,7]. The calibration of mass number was made by using the nuclides ^{143}Sm^{m} which was produced in the ^{141}Pr(^{6}Li,4n) reaction, ^{237}Am, and ^{221}Fr recoiling out of the ^{225}Ac α source. The mass resolution of about 600 ($A/\Delta A$) was achieved for the isotope ^{221}Fr at the focal plane. Mass-separated products were collected on an aluminum coated Mylar tape in a tape transport system and periodically transported to a measuring position having Mylar windows on both sides. The detection system was equipped with a planar-type Ge detector [51 mm diameter×20 mm thick, full width at half maximum (FWHM)=0.57 keV at 122 keV] and a 36% coaxial n-type HPGe detector (FWHM=1.8 keV at 1.33 MeV). Both detectors were used for the X/γ-ray singles and γ-γ coincidence measurements. The data were recorded event by event together with time information.

Figure 1 shows a γ-ray spectrum obtained at the mass-237 fraction accumulated during the period of 6400 s×8 cycles. Pu$K_{\alpha1}$ and $K_{\alpha2}$ x rays and a 280.3 keV γ ray associated with the EC decay of ^{237}Am are seen and x-γ coincidence events between Pu K x rays and a 280.3 keV γ ray were observed. The average half-life of these x- and γ-ray

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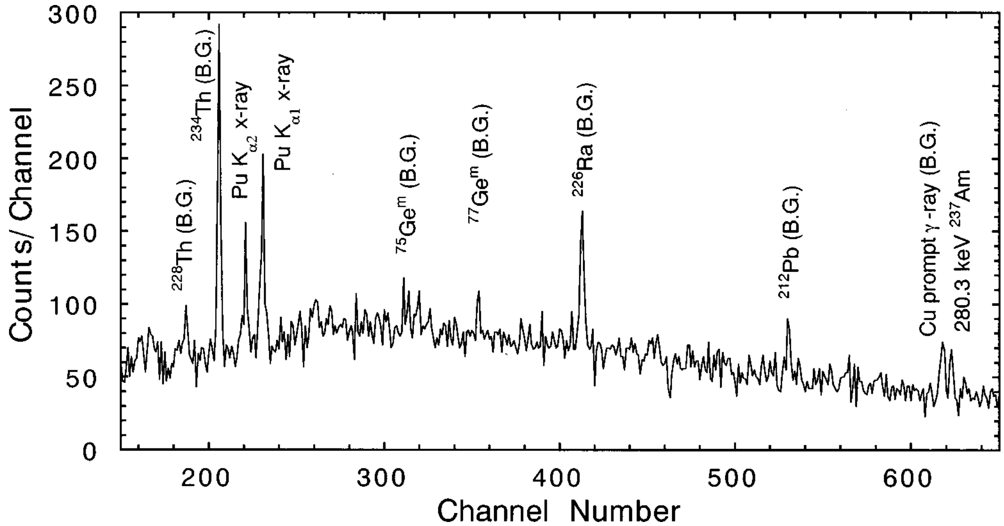

FIG. 1. γ-ray spectrum observed at the mass-237 fraction.

intensities is 1.2 ± 0.5 h which is consistent with the half-life of ^{237}Am [9]. Thus, it is ascertained that the americium atoms [ionization potential (IP)=5.99 eV] are successfully ionized and mass separated with the present system. As the average production cross section of the ^{235}U(^{6}Li,4n)^{237}Am reaction for the present energy range is estimated to be about 250 μb [10], the overall efficiency including the ionization is approximately evaluated to be 0.1%. At the mass-237 fraction, there are no γ rays originating from the decay of ^{238}Am, so that the contamination of ^{238}Am is estimated to be less than 1% based on the counting statistics.

A γ-ray spectrum observed at the mass-236 fraction is given in Fig. 2. The data were accumulated during 400 s $\times$230 cycles and 640 s$\times$200 cycles. Pu$K_{\alpha1}$ and $K_{\alpha2}$ x rays are clearly seen and the Pu$K_{\beta1,3}$ x ray is also observed, while the other γ rays are assigned to the background. No γ-γ coincidence events were detected within the statistics. The contribution from other americium isotopes to the present PuK x rays can be considered to be negligible not only from

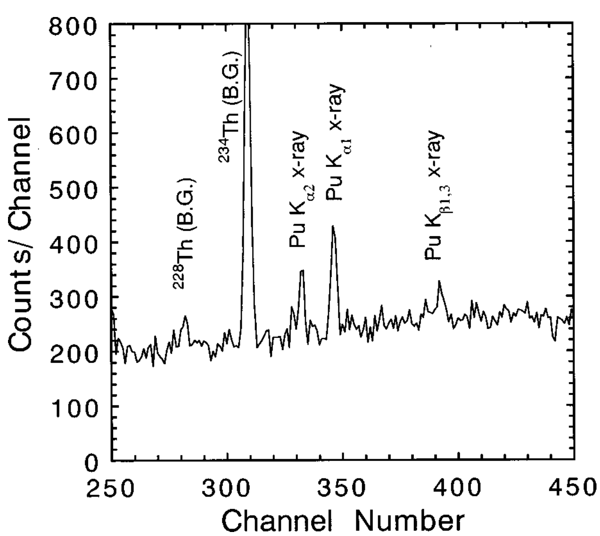

FIG. 2. γ-ray spectrum observed at the mass-236 fraction.

the mass resolution of this system, but also from the facts that (i) no γ rays from ^{237}Am are observed, and (ii) according to the statistical model calculations using the ALICE code [11], the production yield of ^{235}Am via the ^{235}U(^{6}Li,6n) reaction with 46–50 MeV ^{6}Li ions is estimated to be less than about two orders of magnitude of that of ^{236}Am via the ^{235}U(^{6}Li,5 n) reaction. Concerning the contribution from the isobar, there is little possibility of the existence of an isomeric state of the even-even daughter nuclide ^{236}Pu. It should be noted that according to the known decay properties of ^{236}Np^{m} [12] there is no probability of PuK x rays being emitted following the β^- decay of ^{236}Np^{m} ($t_{1/2}$=22.5 h) which might be produced in the nucleon transfer reactions. Furthermore, it is difficult by the present ion source to ionize neptunium atoms that have such a high ionization potential as IP=6.27 eV. Such considerations lead to a judgement that the contribution to the PuK x rays arising from the β^- decay of ^{236}Np^{m} is also negligible. From these facts, we conclude that the PuK x rays in Fig. 2 are solely ascribable to the EC decay of ^{236}Am. The decay curves of the Pu$K_{\alpha1}$ and $K_{\alpha2}$ x-ray intensities are shown in Fig. 3, and each half-life determined is 4.4 ± 1.0 and 4.5 ± 1.5 min, respectively. Thus the half-life of ^{236}Am is evaluated to be 4.4 ± 0.8 min.

The preliminary half-life and decay properties of ^{236}Am were given by Hall *et al.* who produced ^{236}Am via the reaction of ^{237}Np(^{3}He,4n) [5]. After chemical separation of americium fraction, they observed the Pu$K_{\alpha1,2}$ and $K_{\beta1}$ x rays associated with the EC decay of americium isotopes in γ-ray spectrometry and analyzed the two-component decay curve of the PuK x-ray peaks. It was indicated that the long component of the decay curve was attributable to a mixture of those from ^{237}Am and ^{238}Am, while the short component of that, 3.73 ± 0.28 min, was assigned to ^{236}Am. They estimated that there could be a maximum of about 15% as much ^{235}Am as ^{236}Am produced in the energy range studied. Hall *et al.* also obtained the α-decay branching ratio as $(4.2\pm0.6)\times10^{-4}$ and the upper limit on the probability of ECDF, 2.5×10^{-8} [5]. Although there are some ambiguities in the

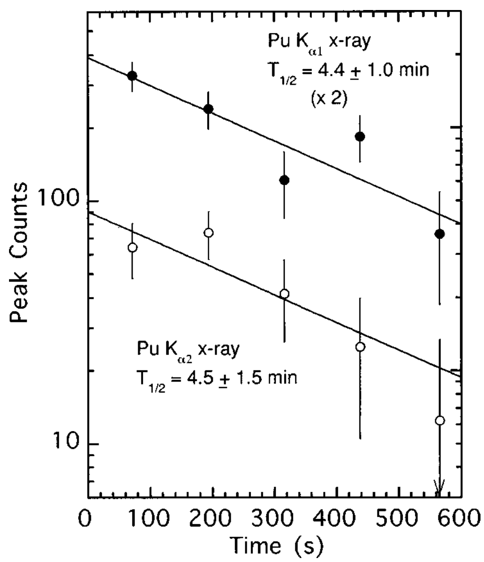

FIG. 3. Decay curve for the Pu$K\alpha_{1,2}$ x rays.

half-life determination by Hall *et al.*, the present result is in agreement with their value [5] within the error limit. Taking the results of Ref. [5] into account, it is concluded that ^{236}Am decays via EC($\sim$100%) with the half-life of 4.4$\pm$0.8 min and the average production cross section of the ^{235}U(^{6}Li,5n)^{236}Am reaction with 46–50 MeV is about 70 μb.

The present result is compared in Table I with the theoretical predictions calculated with the gross theory [13] and the microscopic pn-QRPA (proton-neutron quasiparticle random phase approximation) [14] together with the result of Ref. [5]. The present half-life is shorter than the calculated ones by a factor of about 2. As the calculated values depend on the Q_{EC} values, the discrepancy between the experimental and theoretical would become small if the recent Q_{EC} value of 3.34 MeV [17] is used.

In the following, the degree of forbiddeness is discussed for the observed EC decay of ^{236}Am. The spin and parity of the proton (Z=95) and neutron (N=141) of ^{236}Am are expected to be 5/2$^-$ [523] and 5/2$^+$ [633], respectively, based on the systematics of the spin and parity of the known Am isotopes ^{241,243}Am and those of the isotones ^{233}U and ^{231}Th. Thus the ground state spin-parity of ^{236}Am is expected to be 0$^-$ or 5$^-$. If there are some EC transitions from a certain state of ^{236}Am to the excited levels of ^{236}Pu, γ transitions between the rotational bands, such as 158.4 and 102.8 keV should be observed. However, no γ-ray and no Pu x-γ coincidence was observed as mentioned. Therefore, the observed EC decay of ^{236}Am is considered to be the first forbidden (nonunique) transition from the 0$^-$ state to the ground state of ^{236}Pu with ΔI=0 and $\Delta\pi$=1, i.e., $0^- \to 0^+$. As the probabilities of α and fission decays are negligible [5], the observed EC decay half-life yields a logft of 5.4$\pm$0.1 [18] by using the Q_{EC}=3.34 MeV [17]. Figures 4(a) and 4(b) summarize the experimental logft values for the allowed and first forbidden transitions in the heavy mass region A=231–250, respectively. The logft value of 5.4 $\pm$0.1 is slightly smaller than those reported for the first forbidden transition in this mass region. However, the present result is consistent with the prediction of the gross theory by Yamada *et al.*, according to which the first forbidden transition becomes important and competes with the allowed transition in β decays of heavy nuclei [19]. It is to be added that the existence of the isomeric state with 5$^-$ in ^{236}Am is probable. In fact, the possibility of a long-lived isomeric state in ^{236}Am was suggested by Marinov *et al.* [20].

TABLE I. The measured half-life values of ^{236}Am compared with the results of the two theoretical predictions.

Present result	Hall [5]	Gross theory [13][a]	pn-QRPA [14][b]
4.4$\pm$0.8 min	3.73$\pm$0.28 min	11.1 min	8.87 min

[a] Q_{EC}=3.28 MeV [15].
[b] Q_{EC}=3.13$\pm$0.14 MeV [16].

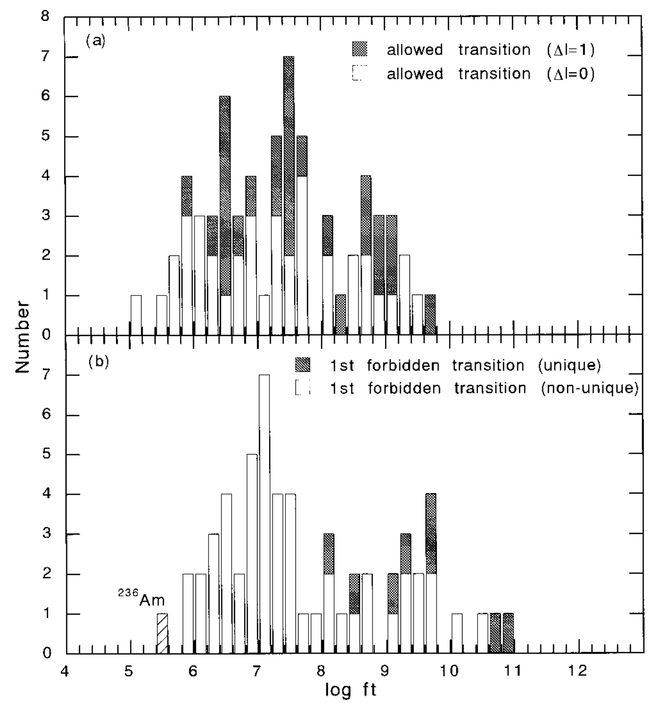

FIG. 4. Experimental logft values in the (a) allowed and (b) first forbidden transitions in the mass region A=231–250.

In conclusion, americium atoms have been successfully ionized and mass-separated with the gas-jet coupled JAERI-ISOL system, and the neutron deficient americium isotope ^{236}Am produced in the reaction of ^{235}U(^{6}Li,5n) was identified. The half-life of ^{236}Am has been measured to be 4.4 $\pm$0.8 min, and this transition is deduced to be the first forbidden (nonunique) EC decay from the 0$^-$ state of ^{236}Am to the ground state ^{236}Pu. The logft value of this transition was evaluated to be 5.4$\pm$0.1.

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