Decay Studies of Neutron-deficient Am, Cm, and Bk Nuclei Using an On-line Isotope Separator

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The EC and α decays of neutron-deficient Am and Cm nuclei have been studied using a gas-jet coupled on-line isotope separator. Decay schemes of the EC decay of ^{235,236}Am have been constructed, and weak α decays of ^{233,235,236}Am and ^{237,238}Cm have been observed. The efficiency of the on-line mass separation of Bk nuclei was measured to be ∼1%. The Q_α values, α-decay partial half-lives, and proton-neutron configurations are discussed.

1. Introduction

Spectroscopic studies of heavy and superheavy nuclei give us information on various nuclear properties in heavy element region. Measured α-decay energies lead to the precise determination of atomic masses which are one of the most essential quantities to define nuclear decay properties, fission and fusion probabilities, and shell stability in heavy nuclei. Gamma-ray spectroscopy following α, β, and orbital-electron capture (EC) decays and fine structure of α-particle spectra reveal excited states in daughter nuclei, single-particle energies, proton-neutron configurations, and nuclear deformation.

Neutron-deficient Am, Cm, and Bk nuclei predominantly decay by EC, and their α-decay probabilities are very small. This property makes experimental studies for these nuclei difficult. In order to measure γ rays following the EC decay and very weak α transitions, it is quite important to isolate the nuclei of interest from a large amount of other reaction products. The traditional method to separate heavy nuclei is chemical separation owing to its high separation efficiency. Another efficient method is in-flight separation using recoil separators. However, these methods are not so suitable to apply to the neutron-deficient Am, Cm, and Bk nuclei; their half-lives are in the order of a few minutes, and their α-decay intensities are very small. In this work, we have studied EC and α decays of the neutron-deficient Am and Cm nuclei using the gas-jet coupled JAERI on-line isotope separator (ISOL)[1] and also succeeded in the on-line mass separation of Bk nuclei. Extremely low-contaminated α/γ sources from the ISOL enabled us to observe weak α/γ transitions with unambiguous mass identification.

2. Experiments

Neutron-deficient Am and Cm nuclei were produced by the reactions of ^{233}U(^{6}Li, xn)$^{233-235}$Am, ^{235}U(^{6}Li, 5n)^{236}Am, and ^{237}Np(^{6}Li, xn)^{237,238}Cm at the JAERI tandem accelerator facility. A stack of twenty-one uranium or neptunium targets set in a multiple-target chamber with 5 mm spacings was bombarded with a ^{6}Li beam of about 400 particle-nA intensity. Each target was electrodeposited with an effective thickness of about 100 μg/cm^2. Reaction products recoiling out of the targets were stopped in He gas loaded with PbI_2 clusters, and transported into an ion source of the ISOL with gas-jet stream through an 8 m long capillary. Atoms ionized in the surface ionization-type thermal ion source were accelerated with 30 kV and mass-separated with a resolution of $M/\Delta M \sim 800$. The overall efficiency of this ISOL system was measured to be 0.3% for ^{237}Am produced in the ^{235}U(^{6}Li, 4n) reaction.[2]

For γ-ray measurements, the separated ions were implanted into an aluminum-coated Mylar tape in a tape transport system, and periodically transported to a measuring position equipped with two Ge detectors. Gamma-ray singles, γ-γ coincidence, and γ-γ delayed coincidence measurements were performed. For α-decay measurements, the separated ions were implanted into 10 μg/cm^2 thick PVC/PVAc foils set on the periphery of a four-position rotating wheel. The wheel periodically rotates 90° to convey the implanted sources to three consecutive detector stations. Each of the detector stations was equipped with two Si

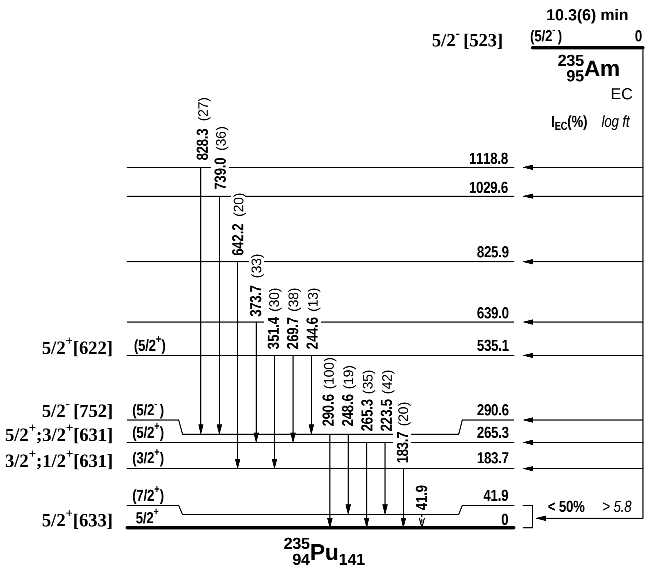

Figure 1. A partial decay scheme of the EC decay of ^{235}Am.

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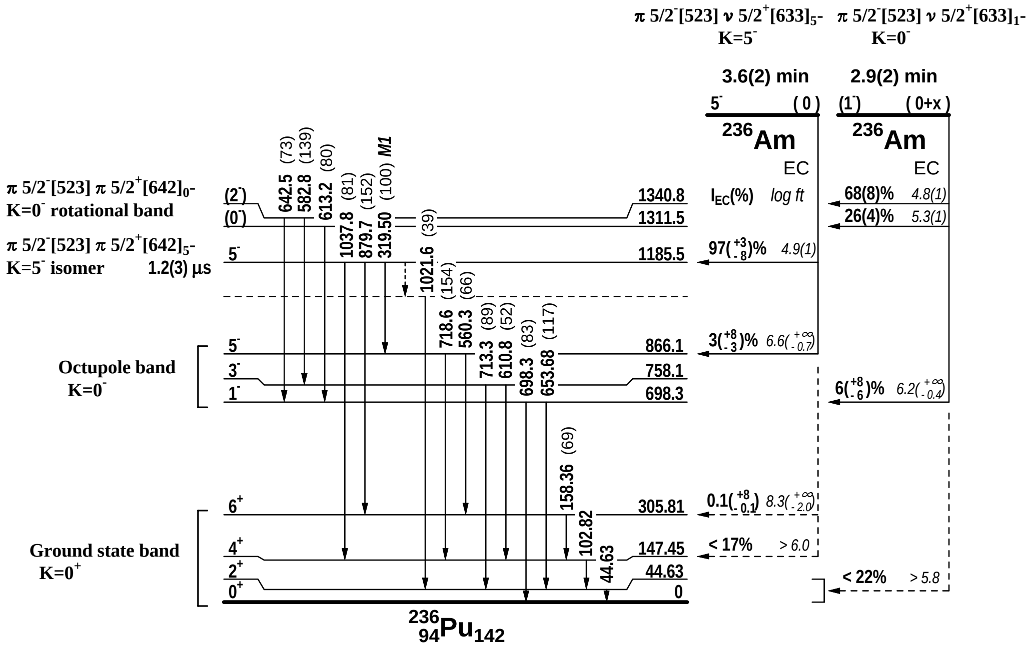

Figure 2. A decay scheme of the EC decay of ^{236}Am.

detectors placed on both sides of the foil to measure α particles with 85% efficiency. An additional Si detector was placed at the implantation station just behind the foil. To determine α/EC branching ratios, a Ge detector was placed at the first detector station. Alpha and γ-ray singles, and α-γ coincidence measurements were performed. The α decay of ^{237}Cm was also measured with another experimental setup where the separated ions were implanted directly into a Si detector. For ^{238}Cm, the tape transport system equipped with seven Si detectors was used to measure its half-life. Details of the experimental procedures are given in Reference 3 and in a forthcoming paper.

3. Results and Discussions

3.1. EC Decay of ^{235,236}Am. Figure 1 shows a partial decay scheme of ^{235}Am established on the basis of the observed γ-γ coincidence relationships. The <50% EC-branching intensity to the ground state of ^{235}Pu was deduced from the comparison between the observed Pu KX-ray intensity and the calculated one based on the proposed decay scheme.

According to the energy systematics of Nilsson single-particle states, the 95th proton of the ground state of ^{235}Am is expected to occupy either the π5/2$^-$[523] orbital or the π5/2$^+$[642] one. Around the neutron-deficient Am region, most of EC and β$^-$ transitions show $\log ft \geq 5.8$ except for the transitions between the π5/2$^+$[642] and the ν5/2$^+$[633] orbitals whose $\log ft$ values are $5.2 \sim 5.4$.[4] If the configuration of the ground state of ^{235}Am is the π5/2$^+$[642], the EC transition to the ν5/2$^+$[633] ground state in ^{235}Pu should have a small $\log ft$ value of ∼5.3. The deduced $\log ft$ value of >5.8 therefore suggests that the π5/2$^+$[642] assignment is unlikely for the ground state of ^{235}Am.

The EC decay of $^{237-240}$Am is characterized by the intense π5/2$^-$[523] → ν5/2$^+$[622] transition with $\log ft = 6.0$–6.2.[5–8] The observed 535 keV level in ^{235}Pu is a candidate for the ν5/2$^+$[622] state because of the large EC feeding to this level. The 41.9 keV level in ^{235}Pu is most likely the 7/2$^+$ state in the 5/2$^+$[633] band. The 291, 265, and 184 keV levels are considered to be the 5/2$^-$[752] band head, the 5/2$^+$ state in the 3/2$^+$[631] band, and the 3/2$^+$ state in the 1/2$^+$[631] band, respectively, owing to the γ-ray branching ratios from these levels to the 5/2$^+$[633] band compared with those in the $N = 141$ isotones.

Figure 2 shows a decay scheme of the EC decay of ^{236}Am. Only the ground state band with spins up to 16$^+$ has been known in ^{236}Pu (Ref. 4). The 698, 758, and 866 keV levels are considered to be the 1$^-$, 3$^-$, and 5$^-$ states in the $K^\pi = 0^-$ octupole band which typically appears at low excitation energy in light actinide nuclei. The energy spacings and γ-ray branching ratios are consistent with this assignment. From the KX to γ intensity ratio, the K-internal conversion coefficient of the 320 keV transition was deduced to be $\alpha_K = 0.87(21)$. This reveals that the 320 keV transition has an M1 multipolarity, and thus, the spin-parity of 5$^-$ is assigned to the 1186 keV level. It has been also found that the 1186 keV level is a K isomer with $K^\pi = 5^-$ and $t_{1/2} = 1.2(3)$ µs. Decay curves of γ rays have revealed that there are two EC-decaying states in ^{236}Am; one is a high-spin state with $T_{1/2} = 3.6(2)$ min, and the other is a low-spin one with $T_{1/2} = 2.9(2)$ min.

The EC transitions from ^{236}Am to the 1186, 1312, and 1341 keV levels in ^{236}Pu show small $\log ft$ values of 4.9, 5.3, and 4.8, indicating that the π5/2$^+$[642] → ν5/2$^+$[633] transition largely contributes to these transitions, that is, the configuration of the ^{236g,m}Am must include either the π5/2$^+$[642] or the ν5/2$^+$[633] orbital. Since the transition to the 1186 keV 5$^-$ state is allowed, the EC-decaying high-spin state has an odd parity. These restrictions in conjunction with the energy systematics of Nilsson single-particle states in $Z = 95$ and $N = 141$ nuclei lead to the (π5/2$^-$[523]ν5/2$^+$[633])5$^-$ assignment for the EC-decaying high-spin state, and this 5$^-$ state is considered to be the ground state according to the Gallagher and Moszkowski coupling rule.[9] The candidate for the low-spin isomer is the 0$^-$ state or its signature partner 1$^-$ state with $K^\pi = 0^-$ and the π5/2$^-$[523]ν5/2$^+$[633] configuration.

Since the ^{236g,m}Am have the π5/2$^-$[523]ν5/2$^+$[633] configuration and the occupied π5/2$^+$[642] orbital, the π5/2$^+$[642] → ν5/2$^+$[633] transition generates the π5/2$^-$[523]π5/2$^+$[642] configuration, hence the 1186, 1312, and 1341 keV states in ^{236}Pu are assigned to be the π5/2$^-$[523]π5/2$^+$[642] two-quasiparticle states. The π5/2$^-$[523]π5/2$^+$[642] two-quasiparticle states were also reported in ^{240}Pu at 1309, 1411, and 1438 keV with spin-parities of 5$^-$, 0$^-$, and 2$^-$, respectively.[4] The 1309 keV level is also the $K^\pi = 5^-$ isomer, and the 1411 and 1438 keV levels are interpreted as the $K^\pi = 0^-$ (π5/2$^-$[523]π5/2$^+$[642])0$^-$ state and its 2$^-$ rotational band member, respectively.[10] If the 1312 and 1341 keV levels in ^{236}Pu are the same 0$^-$ and 2$^-$ states, the ^{236m}Am is assigned to be the 1$^-$ state with $K^\pi = 0^-$. The intensity ratio between the EC transitions from ^{236m}Am to the 1312 and 1341 keV levels is consistent with this interpretation.[11]

3.2. Alpha Decay of $^{233-236}$Am. The α decay of ^{233}Am was unambiguously identified through the observation of the α-α correlations between the α decay of ^{233}Am and following five-successive α decays of ^{229}Np→^{225}Pa→^{221}Ac→^{217}Fr→^{213}At→^{209}Bi. The observed α-particle energy for ^{233}Am was 6780(17) keV, and the half-life of ^{233}Am was determined to be 3.2(8) min from the decay curve of the α particles. Pu KX rays associated with the EC decay of ^{233}Am were not observed. Taking account of the detection efficiency for the Pu KX rays, the α-branching intensity was estimated to be $I_\alpha > 3\%$.[3]

For the α decay of ^{234}Am, Hall et al.[12] reported a 6.46 MeV α group with an α/EC branching ratio of $3.9(12) \times 10^{-4}$ and a 2.32(8) min half-life determined from the decay curve of EC-delayed fission events. In the present experiment, Pu KX-ray peaks were weakly observed in the mass-234 fraction, but no α peak was observed at 6.46 MeV; the upper limit of $I_\alpha < 0.04\%$ was deduced for the 6.46 MeV α group.

In the mass-235 fraction, the 6457(14) keV α peak whose half-life was in good agreement with that of the Pu K_αX rays was clearly observed. The deduced half-life value of 10.3(6) min is consistent with the literature value of 15(5) min by Guo et al.[13] extracted from the growth and decay of ^{235}Pu in chemically purified Am fractions. The α-branching intensity of $I_\alpha = 0.40(5)\%$ was derived from the ratio between the observed α and Pu KX-ray intensities. No α-γ coincidence event was observed for the 6457 keV peak, indicating that the upper limit of the expected E1 transition energy between the $5/2^-$[523] state and the $5/2^+$[633] ground state in ^{231}Np should be <15 keV.

Hall[14] reported a 6.41 MeV α group with an α/EC branching ratio of $4.2(6) \times 10^{-4}$ for ^{236}Am. In the present experiment, however, no 6.41 MeV α peak was observed; the upper limit of the branching intensity for the 6.41 MeV α particles was $I_\alpha < 0.002\%$. Instead of it, a very weak α group with a few-minute half-life and $I_\alpha = 0.004\%$ was observed at 6150 keV. We tentatively assign this α group to that associated with ^{236}Am.

Figure 3 shows α-decay partial half-lives $T^{\alpha}_{1/2}$ for the ground state to ground state transitions in even-even Pu and Cm isotopes and those for the favored transitions in odd-mass $^{237-243}$Am isotopes as a function of the released α-decay energy E^*_α which is a measured α-particle energy corrected by its recoil energy and electron screening.[15] The $T^{\alpha}_{1/2}$ values of ^{233,235}Am lie on the semi-empirical curve[15] fitted to the values of the favored transitions in $^{237-243}$Am, indicating that the α transitions observed in ^{233,235}Am are the favored transition. Since the ground state of ^{235}Am has the $\pi 5/2^-$[523] configuration, the favored transition populates the $\pi 5/2^-$[523] state in ^{231}Np. The $\pi 5/2^-$[523] state in Np isotopes decreases in energy with decreasing neutron number, 60, 49, and <15 keV in ^{237}Np, ^{235}Np, and ^{231}Np, respectively, corresponding to the decreasing deformation, and in ^{229}Np it is expected to become further lower. Since the uncertainties of the level energies populated by the α decay of ^{233,235}Am are as small as those of the α-particle energies, we can determine the Q_α values of ^{233,235}Am within ±20 keV uncertainties. The deduced Q_α values are by 100–200 keV smaller than the evaluated ones by Audi et al.[16]

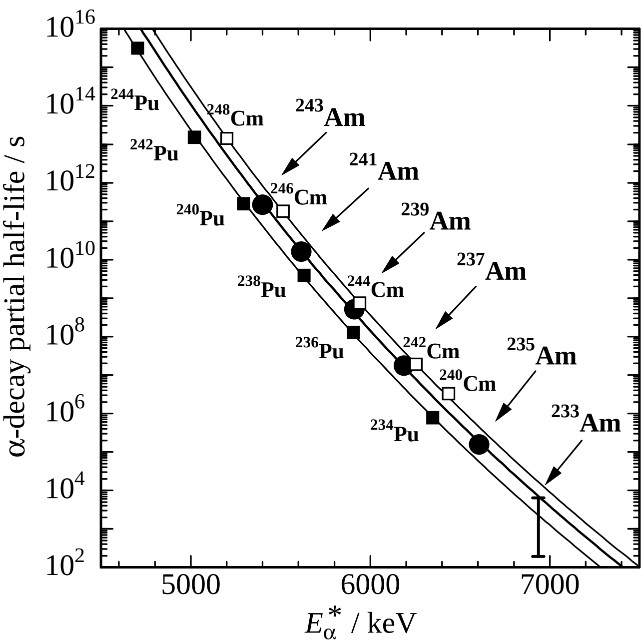

Figure 3. Alpha-decay partial half-lives as a function of the released α-decay energy.

3.3. ^{237}Cm and ^{238}Cm. Figures 4(a) and (b) show α-particle spectra observed in the mass-237 and 238 fractions separated from products in the ^{237}Np(^{6}Li, xn) reaction. The α lines observed at 5500, 5773, and 6047 keV are attributable to ^{238}Pu, ^{236}Pu, and ^{237}Am, respectively. Taking account of the mass resolution of the present ISOL system, about 0.2% of mass-separated ^{236}Pu may be observed in the adjacent mass-237 fraction. The 6660(10) keV line was attributed to the α decay of ^{237}Cm. Its mass identification was confirmed by measuring the adjacent mass fractions. The α-particle energy of ^{238}Cm was revised from the literature value of 6520(50) keV[17] to the present one of 6560(10) keV. The half-life of ^{238}Cm was determined to be 2.2(4) h, while that of ^{237}Cm could not be determined because of its less statistics.

4. Future Plans

Neutron-deficient Bk nuclei have been studied scarcely because of their short half-lives and small α-branching intensities. The ^{238,240}Bk were identified through the observation of EC-delayed fission events,[18,19] and Cm KX rays associated with the EC decay of ^{242}Bk ($T_{1/2} = 7.0$ min) were observed through the chemical separation.[20] In future plans, we intend to measure the EC decay of ^{241}Bk produced in the ^{239}Pu(^{6}Li, $4n$)^{241}Bk reaction using the present ISOL system. In order to check the ionization efficiency of Bk, the on-line mass separation of ^{250}Bk ($T_{1/2} = 3.2$ h) produced in the transfer reaction with a ^{248}Cm target and ^{18}O projectiles has been performed. The intensity of γ rays from the mass-separated ^{250}Bk was compared with that measured by using only the gas-jet transport. The intensity ratio of 2% was obtained. This value does not include only the gas-jet transport efficiency which is about 50% in the present system. Thus, the overall efficiency is estimated to be ∼1% which is comparable to that of Am isotopes. The ionization efficiency depends not only on the ionization potential of each element but also on the vapor pressure. The ionization potential

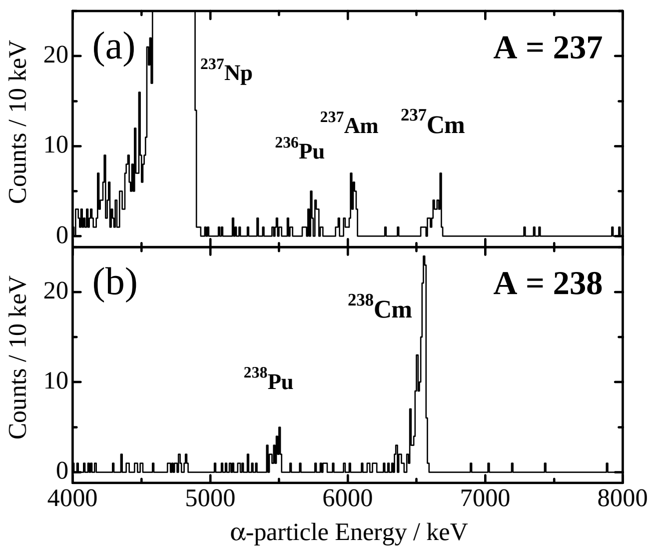

Figure 4. Alpha-particle spectra observed in (a) the mass-237 fraction and (b) the mass-238 fraction.

increases with increasing atomic number from Am to probably No, which makes the ionization efficiency decrease. However, if the vapor pressure increases, the ionization efficiency increases. Therefore, it may be expected that Es, Md, and No isotopes are also ionized efficiently and their EC decays are studied using the present ISOL system.

The other plan is α-γ coincidence measurements for ^{259,261}Rf and ^{257}No to assign spin-parities and Nilsson orbitals in $N > 153$ nuclei. In this experiment, we only use the gas-jet transport without using the ISOL, and γ, KX, LX, and internal conversion electrons in coincidence with α particles are measured with good statistics. Then, we deduce level energies and multipolarities of γ transitions from internal conversion coefficients. In combination with α-particle energies and their hindrance factors, we can extract information on spin-parities and Nilsson orbitals of excited states in daughter nuclei as well as the ground states of the parents. In addition, we can also determine Q_α values without any assumptions on the populated level energies. The nucleus ^{261}Rf is located on the α-decay chain starting from the nucleus ^{277}112. The above spectroscopic data provide an experimental basis to discuss nuclear structure in heavy and superheavy region.

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