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Heavy Element Nuclear Chemistry Research in JAERI *

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Abstract

Heavy element nuclear chemistry research in JAERI is reviewed. Recent experimental results on decay studies of neutron deficient actinide nuclei using the gas-jet coupled JAERI-ISOL are presented. Successful production of the transactinide nuclei, ^{261}Rf and ^{262}Db, and the present status of studies of chemical properties on the transactinide elements are introduced.

1 Introduction

Nuclear chemistry study of heavy elements in JAERI is currently being performed at the JAERI tandem accelerator facility. Nuclear decay studies of neutron deficient actinide nuclei are conducted using the gas-jet coupled JAERI-ISOL system and recently the new isotopes ^{233,236}Am and ^{237}Cm have been successfully identified. Quite recently, the transactinide nuclei ^{261}Rf and ^{262}Db have been produced for the first time in Japan via the reactions of ^{248}Cm(^{18}O,5n) and ^{248}Cm(^{19}F,5n), respectively. In this report, the present status of nuclear chemistry studies of heavy elements in JAERI is briefly summarized.

2 EC/α decay studies of neutron deficient actinides

There still remain many unknown isotopes to be discovered in the region of neutron deficient actinides which predominantly decay through the electron capture (EC). 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.

To search for new isotopes and study EC/α decay properties of neutron deficient actinides, 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) [1]. This gas-jet coupled JAERI-ISOL system enables us to determine simultaneously mass number via the isotope separator and atomic number by the measurement 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

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with this system [2-5]. In the following, the EC/α decay studies of the neutron deficient actinides are summarized.

The experimental setup is schematically drawn in Fig. 1. The ^{6}Li beams delivered from the JAERI tandem accelerator bombarded the ^{233,235}U and ^{237}Np targets set in a multiple target chamber. The used reaction systems are ^{233}U(^{6}Li,xn)$^{233-235}$Am, ^{235}U(^{6}Li,5n)^{236}Am and ^{237}Np(^{6}Li,xn)^{237,238}Cm. Reaction products recoil out of the targets were thermalized in He gas loaded with PbI_2 aerosol clusters. The products attached to the aerosols were swept out of the target chamber and transported to the thermal ion source of ISOL through a capillary (1.5 mm i.d. and 8 m length). The transported nuclides were ionized in the thermal ion source, and mass-separated atoms were collected on an aluminum coated Mylar tape in a tape transport system or a rotating catcher foil apparatus. In the tape transport system, we use Ge detectors for the x/γ-ray measurement and the Si photodiode detectors set in the rotating catcher foil apparatus were employed for the measurement of α-rays. Detailed experimental procedures are described in [6, 7].

As a typical example, the identification of the new isotope ^{233}Am produced in the ^{233}U(^{6}Li,6n) reaction is described [7]. The isotope ^{233}Am was identified through an $\alpha-\alpha$ correlation analysis. The α decay of ^{233}Am is followed by the five successive α decays starting from ^{229}Np as shown in Fig. 2: ^{229}Np ($t_{1/2}$=4.0 min) $\rightarrow$ ^{225}Pa ($t_{1/2}$=1.7 s) $\rightarrow$ ^{221}Ac ($t_{1/2}$=52 ms) $\rightarrow$ ^{217}Fr ($t_{1/2}$=22 μs) $\rightarrow$ ^{213}At ($t_{1/2}$=125 ns) $\rightarrow$ ^{209}Bi (stable). Since the last four nuclides decay via α particle emission with the short half-lives, the α-α correlation events among these nuclides can be unambiguously identified. Figure 2 shows an α-particle spectrum constructed from the observed α-α correlation events in the mass-separated A=233 fraction. The α-ray energies of ^{233}Am and ^{229}Np are clearly observed and those corresponding to the decays of the other nuclides are also seen. The α-ray energy of ^{233}Am was determined to be 6780$\pm$17 keV, and from the decay curve of the intensity of this α-line, the half-life was 3.2$\pm$0.8 min. Since no Pu Kx-rays following the EC-decay of ^{233}Am was observed, the α-decay branching ratio was estimated as I_α >3% based on the detection efficiency of the Pu Kx-rays.

In Table 1, the half-lives, α decay energies and α-decay branching ratios measured in the present study and those compared with the literature data are shown. In the case of ^{235}Am, we first observed the α-decay process and the α branching intensity was derived from the ratio between the observed α and Pu Kx-ray intensities. The half-life value of ^{235}Am has been determined to be 10.3$\pm$0.6 min based on the decay curve of the α line of 6457 keV and that of Pu K_αx-rays following the EC decay of ^{235}Am. With the γ-γ coincidence technique, it is found that there are two EC-decaying states in ^{236}Am [8]. The α-decay processes in ^{234}Am and ^{236}Am were not observed in the preset experiments, although those were reported by Hall *et al.* [9] as shown in Table 1.

Table 1. EC/α decay properties of the neutron-deficient Am and Cm isotopes measured in the present work and those compared with the literature data.

Nuclide	Half-life (min)		α-energy (keV)		α branching ratio (%)	
	Present	Ref.	Present	Ref.	Present	Ref.
^{233}Am	3.2$\pm$0.8	(1.72)*	6780$\pm$17	(7100)*	> 3	
^{234}Am		2.32$\pm$0.08	-	6460	< 0.04	0.039$\pm$0.012
^{235}Am	10.3$\pm$0.6	15$\pm$5	6457$\pm$14	6700	0.40$\pm$0.05	
^{236g}Am	3.6$\pm$0.2	4.4$\pm$0.8	6150	6410	< 0.004	0.042$\pm$0.006
^{236m}Am	2.9$\pm$0.2					
^{237}Cm	$\approx$10	(15)*	6660$\pm$10	(6800)*		
^{238}Cm			6560$\pm$10	6520$\pm$50		

* Predicted

3 Nuclear chemistry studies of the transactinide elements

Studies of chemical properties of the transactinide elements - starting with element 104 (Rf) - offer the unique opportunity to obtain information about trends in the Periodic Table at the limits of nuclear stability and to assess the magnitude of the influence of relativistic effects on chemical properties. According to the calculations of the electron configurations of the transactinide elements, it is predicted that sudden changes in the structure of electron shells may appear due to the relativistic effects which originate from the increasing strong Coulomb field of the highly charged atomic nucleus. Thus, it is expected that the transactinide elements show a drastic rearrangement of electrons in their atomic ground states and as the electron configuration is responsible for the chemical behavior of the element, such relativistic effects can lead to surprising chemical properties. Increasing deviations from the periodicity of chemical properties based on extrapolation from lighter homologues in the Periodic Table are predicted. The main objective of the present group is to explore experimentally the influence of the relativistic effects on the electron shell structure by studying the chemical properties of the transactinide elements. The comparison studies of such fundamental chemical properties as the most stable oxidation states, complex ability and ionic radii with those of lighter homologues, and with results of relativistic molecular orbital calculations are invaluable in evaluating the role of the relativistic effects.

The transactinide elements must be produced by bombarding heavy radioactive actinide targets with high-intensity heavy ion beams and must be identified by the measurement of their decay or that of their known daughter nuclei with unambiguous detection techniques. Detailed studies of nuclear decay properties of transactinide nuclides are also the important subject.

Since 1998, we have developed some experimental apparatus for the transactinide nuclear chemistry study, *i.e.* a beam-line safety system for the usage of the gas-jet coupled radioactive ^{248}Cm target chamber, a rotating wheel catcher apparatus for the measurement of the α and SF decays of the transactinides and an automated rapid chemical separation apparatus based on the high performance liquid chromatography.

Quite recently, we have successfully produced the transactinides, ^{261}Rf and ^{262}Db, by using the ^{248}Cm(^{18}O,5n) and ^{248}Cm(^{19}F,5n) reactions, respectively. Figure 3 shows the schematic of the experiment for the production and identification of ^{261}Rf and ^{262}Db: the target chamber coupled to the gas-jet transport and the rotating wheel catcher apparatus. The ^{248}Cm target of 590 μg/cm^2 thickness was bombarded by the ^{18}O and ^{19}F beams with the intensity of 200-300 pnA. The recoiling products were stopped in He gas, attached to a KCl aerosol, and were continuously transported through a Teflon capillary to the rotating wheel catcher apparatus. The transported nuclei were deposited on polypropylene foils of 240 μg/cm^2 thickness and 20 mm diameter at the periphery of an 80-position stainless steel wheel of 80 cm diameter. The wheel was stepped at 30 s time intervals to position the foils between six pairs of Si PIN photodiode detectors. The details of the experimental procedures are described elsewhere [10].

The sum of α-particle spectra measured in the six top detectors in a 3.9 h irradiation for the production of ^{261}Rf is shown in Fig. 4(a). In the α energy range of 8.12-8.36 MeV, α lines from 78-s ^{262}Rf (8.28 MeV) and its daughter 26-s ^{257}No (8.22, 8.27, 8.32 MeV) are clearly shown. No contributions from other nuclides in this energy window are observed, although there exit several α lines originating from the Pb impurities in the ^{248}Cm target. A total of 98 events in the singles measurement and 28 α-α correlation events were registered. The production cross section of ^{261}Rf in this reaction was evaluated to be about 6 nb at the ^{18}O energy of 99 MeV.

Figure 4(b) shows the sum of α-particle spectra for the production of ^{262}Db in the 100 MeV ^{19}F-induced reaction of ^{248}Cm. From the mother-daughter correlation of α-energies between ^{262}Db-mother and ^{258}Lr-daughter, the cross section of this reaction was about 1 nb.

Because of the short half-lives and the low production rate of the transactinides, each atom produced decays before a new atom is synthesized. This means that any chemistry to be per-

formed must be done on an *atom-at-a-time* basis. Therefore rapid, very efficient and selective chemical procedures are indispensable to isolate the desired transactinides.

We have developed the gas-jet coupled automated chemical separation apparatus to perform rapid, repetitive and high performance liquid chromatography separations on the second time scale. It is equipped with two magazines, each containing twenty micro-columns (1.6 mm in diameter and 8 mm long). A series of chromatographic pumps, valves, mechanical sliders, and micro-columns are all controlled by a personal computer. The performance is almost the same as that of ARCA developed by the GSI-Mainz group [11]. On-line ion exchange experiments with the above apparatus are being carried out using the Rf homologues Zr and Hf produced via the ^{89}Y(p,n)^{89m}Zr and ^{152}Gd(^{18}O,xn)^{165,167}Hf reactions. The Rf chemistry experiments based on the ion exchange behavior will be started in the beginning of 2001.

Acknowledgements

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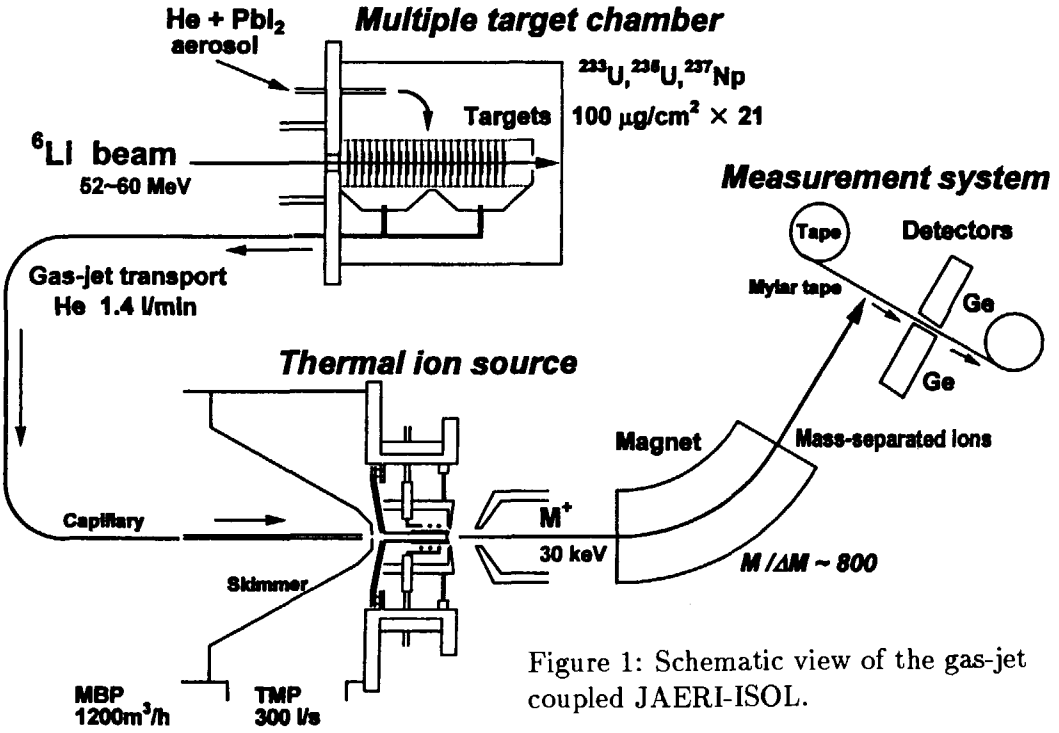

Figure 1: Schematic view of the gas-jet coupled JAERI-ISOL.

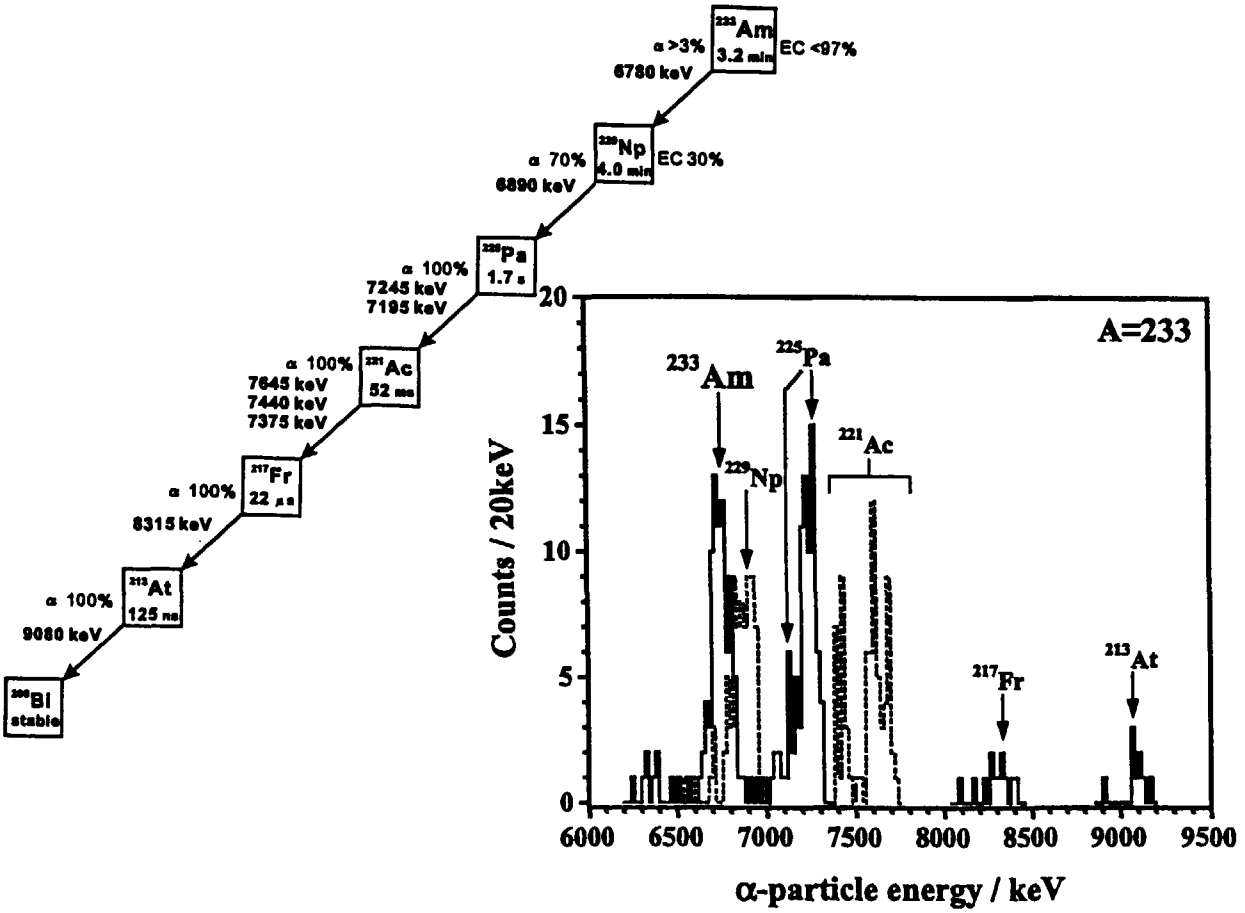

Figure 2: α-decay chain originating from the α decay of ^{233}Am, and α-particle spectrum constructed from the measured α-α correlation events associated with the α-decay of ^{233}Am.

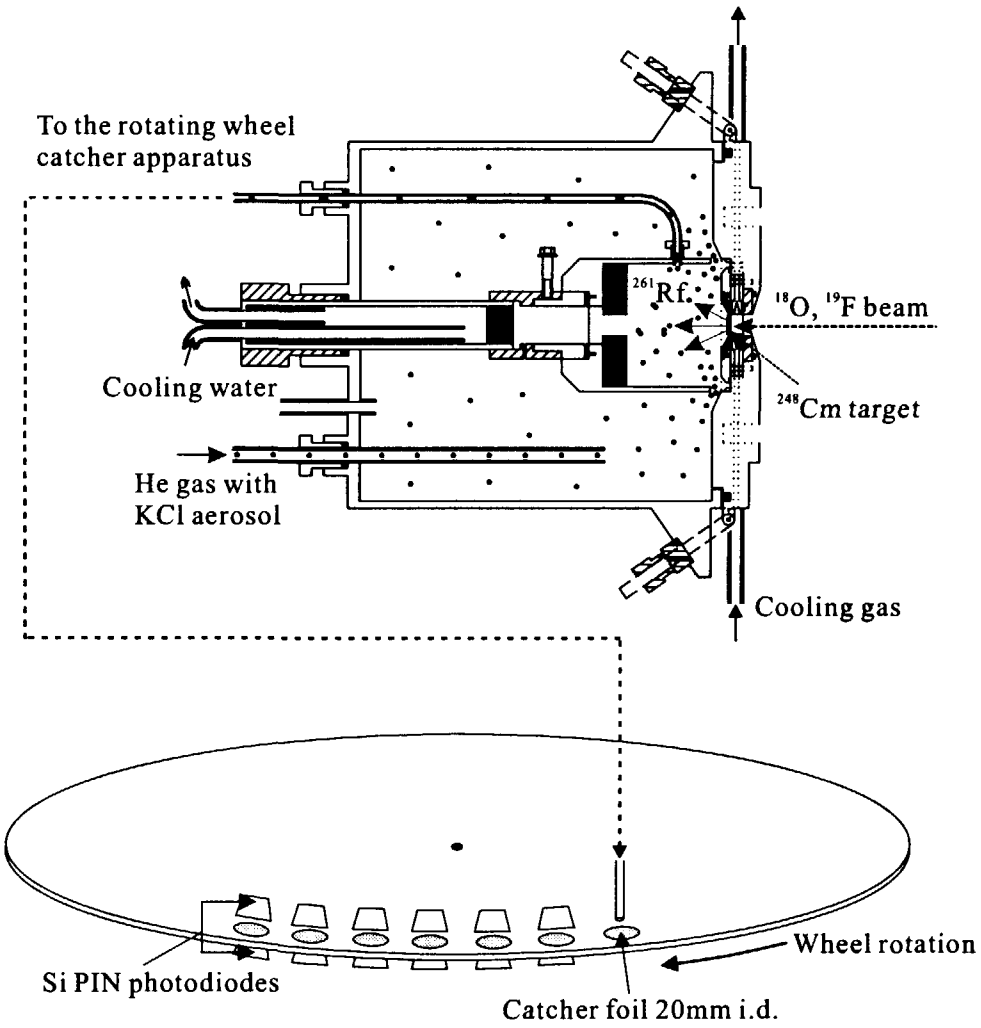

Figure 3: Schematic of the experiment for the production of ^{261}Rf and ^{262}Db.

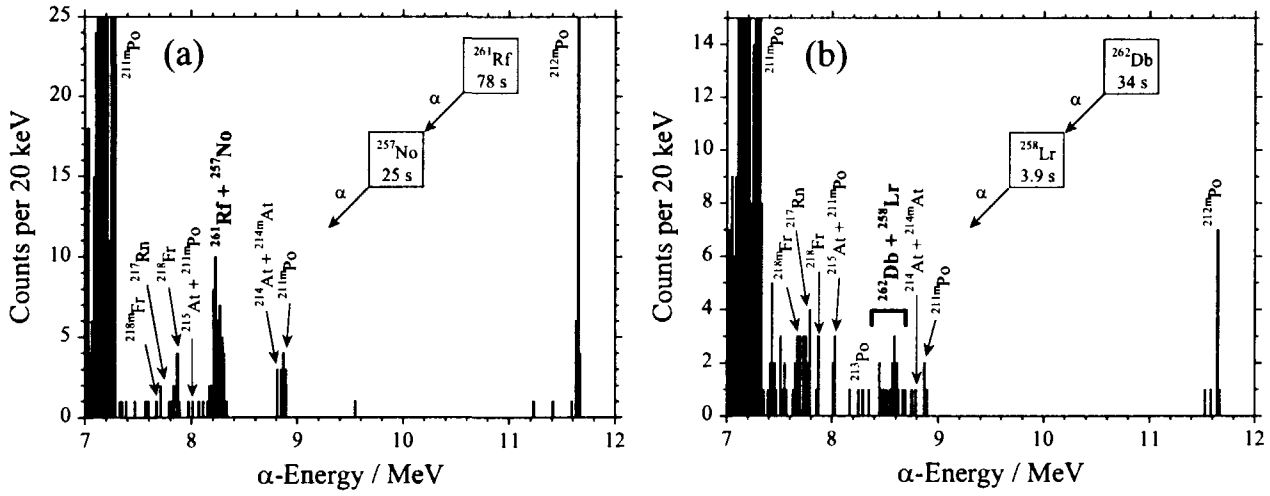

Figure 4: (a) Sum of α-particle spectra measured in the bombardment of the ^{248}Cm target with 99 MeV ^{18}O ions, and (b) that in the bombardment of the ^{248}Cm target with 100 MeV ^{19}F ions.