Status and Prospects of Heavy Element Nuclear Chemistry Research at JAERI

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Received: March 3, 2002; In Final Form: May 9, 2002

The present status of heavy element nuclear chemistry research at JAERI (Japan Atomic Energy Research Institute) is reviewed. Successful production of the transactinide nuclei, ^{261}Rf and ^{262}Db via the reactions of $^{248}\text{Cm}(^{18}\text{O}, 5n)$ and $^{248}\text{Cm}(^{19}\text{F}, 5n)$, respectively at the JAERI tandem accelerator is presented. Aqueous chemistry of Rf is being carried out with a newly developed rapid ion-exchange separation apparatus. Anion-exchange behavior of Rf in HCl solution is shortly discussed. Recent experimental results on decay studies of neutron-deficient actinide nuclei using the gas-jet coupled JAERI-ISOL are also given. Prospects for the near future studies are briefly considered.

1. Introduction

Nuclear chemistry study of the transactinide elements in Japan is currently being performed using the JAERI tandem accelerator. We have developed some experimental apparatuses: a beam-line safety system for the usage of the gas-jet coupled radioactive ^{248}Cm target and recoil chamber, a rotating wheel catcher apparatus for the measurement of α and spontaneous fission decay of transactinides, and an automated rapid ion-exchange separation apparatus based on the high performance liquid chromatography coupled with an on-line α -particle detection system.¹

The schematic of the experimental setup for the study of the transactinide elements is shown in Figure 1. The transactinide nuclei, 78-s ^{261}Rf and 34-s ^{262}Db which are commonly used for recent chemical studies of element 104 and 105, have been successfully produced for the first time in Japan through the reactions of $^{248}\text{Cm}(^{18}\text{O}, 5n)$ and $^{248}\text{Cm}(^{19}\text{F}, 5n)$, respectively. The production cross sections for each reaction have been accurately measured with the newly developed rotating wheel catcher apparatus MANON (Measurement system for Alpha particle and spontaneous fission events ON line).²

On-line anion-exchange experiments of Rf together with the group 4 elements Zr and Hf in HNO_3 and HCl solutions have been conducted with AIDA (Automated Ion exchange separation apparatus coupled with the Detection system for Alpha spectroscopy). The results have clearly shown that ion-exchange behavior of Rf is quite similar to that of Zr and Hf, and is different from that of the tetravalent pseudo-homologue Th, indicating Rf is the member of the group 4 elements.³

Studies of α/EC (electron capture)-decay properties of neutron-deficient actinide nuclei have been carried out using the gas-jet coupled JAERI-ISOL system and recently the new isotopes $^{233,236}\text{Am}$ and ^{237}Cm have been unambiguously identified.^{4–6}

In this report, the present status of nuclear chemistry studies of heavy elements and research plans for the near future at JAERI are summarized.

2. Production of ^{261}Rf and ^{262}Db

As shown in Figure 1, the ^{248}Cm target of $590\text{ }\mu\text{g}/\text{cm}^2$ thickness prepared by electrodeposition onto a $2.2\text{ mg}/\text{cm}^2$ thick beryllium backing foil was bombarded by ^{18}O and ^{19}F beams de-

livered from the JAERI tandem accelerator. The reaction products recoiling out of the target were stopped in He gas ($\sim 1\text{ bar}$), attached to KCl aerosols, and were transported through a Teflon capillary to the rotating wheel apparatus MANON. The transported nuclei were deposited on polyethylene terephthalate foils of $120\text{ }\mu\text{g}/\text{cm}^2$ thickness. The wheel was periodically rotated to position the foils between six pairs of Si PIN photodiodes for α -particle detection. The production cross sections were evaluated from the mother-daughter correlations of α energies between ^{261}Rf and ^{257}No , and ^{262}Db and ^{258}Lr . The experimental details are described in Reference 2.

The typical α -particle spectrum measured in the six top detectors during irradiation of 1.6×10^{17} ^{19}F ions for the production of ^{262}Db in the 106 MeV ^{19}F -induced reaction of ^{248}Cm is shown in Figure 2(a). In the α -energy range of 8.40–8.70 MeV, α lines from 34-s ^{262}Db (8.45, 8.53, and 8.67 MeV) and its daughter 3.9-s ^{258}Lr (8.565, 8.595, 8.621, and 8.654 MeV) are observed. Figure 2(b) shows the mother-daughter (α - α) correlation plot for the 106 MeV $^{19}\text{F} + ^{248}\text{Cm}$ reaction. A total of 30 α - α correlation events were registered. The literature values for the α energies of ^{262}Db and ^{258}Lr are indicated by the solid lines.

From the mother-daughter correlation events, the production cross section of ^{261}Rf was evaluated to be $13 \pm 3\text{ nb}$ at the ^{18}O energy of 94 MeV, while that of ^{262}Db was $1.3 \pm 0.4\text{ nb}$ at 106 MeV ^{19}F (Ref. 2).

3. Aqueous Chemistry of Rf

Study of the chemical properties of the transactinide elements is challenging both experimentally and theoretically.⁷ Due to in-

Figure 1. Schematic drawing of the experimental setup.

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TABLE 1: EC/ α decay properties of the neutron-deficient Am and Cm isotopes measured with JAERI-ISOL and those compared with the literature data.

Nuclide	Half-life / min		α -energy / MeV		α -branching ratio / %	
	JAERI-ISOL	Reference	JAERI-ISOL	Reference	JAERI-ISOL	Reference
^{233}Am	3.2 ± 0.8^6	(1.72) ^a	6.780 ± 0.017^6	(7.100) ^b	$>3^6$	
^{234}Am		2.32 ± 0.08^{18}		6.460^{18}	$<0.04^{16}$	0.039 ± 0.012^{18}
^{235}Am	10.3 ± 0.6^{16}	15 ± 5^{22}	6.457 ± 0.014^{16}	(6.700) ^b	0.40 ± 0.05^{16}	
^{236g}Am	3.6 ± 0.2^{17}	4.4 ± 0.8^5	6.150^{16}	6.41^{19}	0.004^{16}	0.042 ± 0.006^{19}
^{236m}Am	2.9 ± 0.2^{17}					
^{237}Cm		(15) ^a	6.660 ± 0.010^4	(6.800) ^b		
^{238}Cm			6.560 ± 0.010^4	6.520 ± 0.050^{23}		

^aPredicted.²⁰^bEvaluated from the Q values in Reference 21.

other interesting feature was observed in the adsorption trend on the anionic-exchange resin among Rf, Zr, and Hf. The adsorption order that reflects the stability of chloride complexation was $\text{Rf} > \text{Zr} > \text{Hf}$. Presently, we are conducting the relativistic molecular orbital calculations to understand the present results and to see if the anion-exchange behavior of Rf is affected by relativistic effects.

4. 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).^{5,11} 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 with this system.^{12–15} In the following, the EC/ α -decay studies of the neutron-deficient actinides are summarized.

The experimental setup is schematically drawn in Figure 4. The ^6Li beams delivered from the JAERI tandem accelerator bombarded the $^{233,235}\text{U}$ and ^{237}Np targets set in a multiple target chamber. The used reaction systems are $^{233}\text{U}(^6\text{Li}, xn)^{233–235}\text{Am}$,

$^{235}\text{U}(^6\text{Li}, 5n)^{236}\text{Am}$, and $^{237}\text{Np}(^6\text{Li}, xn)^{237,238}\text{Cm}$. The reaction products recoiling 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 Teflon 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 used Ge detectors for the x/ γ -ray measurement and Si PIN photodiode detectors set in the rotating catcher foil apparatus were employed for the measurement of α particles. Detailed experimental procedures are described in References 6, 11.

As a typical example, the identification of the new isotope ^{233}Am produced in the $^{233}\text{U}(^6\text{Li}, 6n)$ reaction is given in the follows. The isotope ^{233}Am was identified through an α - α correlation analysis. The α decay of ^{233}Am is followed by the five successive α decays starting from ^{229}Np as shown in Figure 5: ^{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 6 shows an α -particle spectrum constructed from the observed α - α correlation events in the mass-separated $A = 233$ fraction. The α -particle 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 energy of ^{233}Am was determined to be 6.780 ± 0.017 MeV, and from the decay curve of the intensity of this α line, the half-life was 3.2 ± 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 $I_\alpha > 3\%$ based on the detection efficiency of the Pu Kx rays.⁶

In Table 1, the half-lives, α -decay energies, and α -decay branching ratios measured with JAERI-ISOL and those from the literature data are summarized. In the case of ^{235}Am , the α -decay process was observed for the first time and the α -

Figure 5. α -decay chain originating from the α decay of ^{233}Am . **Figure 6.** α -particle spectrum constructed from the measured α - α correlation events associated with the α decay of ^{233}Am .

branching intensity was derived from the ratio between the observed α and Pu K α -ray intensities.¹⁶ The half-life value of ²³⁵Am was determined to be 10.3 ± 0.6 min based on the decay curve of the α line of 6.457 MeV and that of Pu K α rays following the EC decay of ²³⁵Am. With the γ - γ coincidence technique, it was found that there are two EC-decaying states in ²³⁶Am (Ref. 17). The α -decay process in ²³⁴Am was not observed, although that was reported by Hall et al.¹⁸ as shown in Table 1. In the α decay of ²³⁶Am, a very weak α group was observed at 6.150 MeV, while the 6.41 MeV α group was measured in Reference 19.

5. Future Plans

In the study of Rf aqueous chemistry, anion-exchange experiments in HF solution will be soon conducted. Plans for additional experiments are being made to understand complex structure of Rf together with Zr and Hf in HNO₃, HCl, and HF solutions. Measurements of XAFS (X-ray Absorption Fine Structure) spectra of Zr and Hf compounds in these solutions will be performed at KEK (High Energy Accelerator Research Organization) Photon Factory. Relativistic molecular orbital calculations are also being carried out to gain understanding of complex chemistry of Rf, and to see if there are unexpected differences within the group 4 elements.

In the next stage of aqueous chemistry, we plan to conduct Db ion-exchange experiments. Improvements to the overall production rates and a possible multiple target system are being considered to study the short-lived 34-s ²⁶²Db produced in the ²⁴⁸Cm(¹⁹F, 5n) reaction with the small cross section of about 1.5 nb. Development of a new ion-exchange separation apparatus, an improved AIDA system is required. We are now cooperating with the Niigata group on the gas phase chemistry of the transactinide elements. An apparatus based on the isothermal gas chromatography is now under construction.

Experiments of α - γ spectroscopy, and spontaneous fission and α -decay properties of heavy and transactinide nuclei will be conducted with the gas-jet coupled JAERI-ISOL, an α - γ spectrometry apparatus, and MANON coupled with a gas chemistry separator system.

Acknowledgement. The present study has been carried out in collaboration with Niigata University, Osaka University, Tokyo Metropolitan University, University of Tokushima, Kanazawa University, Nagoya University, Hiroshima University, Gesellschaft für Schwerionenforschung (GSI), Mainz University, Paul Scherrer Institut (PSI), Bern University, and Institute of High Energy Physics, Beijing. We would like to acknowledge the crew of the JAERI tandem accelerator for providing the stable and intense ⁶Li, ¹⁸O, and ¹⁹F beams. This work was partly supported by the JAERI tandem accelerator collaboration program and the Program on the Scientific Cooperation between JAERI and GSI in Research and Development in the Field of Ion Beam Application.

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