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Abstract. A gas-jet transport system has been coupled to the RIKEN gas-filled recoil ion separator GARIS to startup superheavy element (SHE) chemistry at RIKEN. The performance of the system was appraised using an isotope of element 104, ^{261}Rf, produced in the ^{248}Cm(^{18}O,5n)^{261}Rf reaction. Alpha-particles of ^{261}Rf separated with GARIS and extracted to a chemistry laboratory were successfully identified with a rotating wheel apparatus for α spectrometry. The setting parameters such as the magnetic field of the separator and the gas-jet conditions were optimized. The present results suggest that the GARIS/gas-jet system is a promising approach for exploring new frontiers in SHE chemistry: (i) the background radioactivities of unwanted reaction products are strongly suppressed, (ii) the intense beam is absent in the gas-jet chamber and hence high gas-jet efficiency is achieved, and (iii) the beam-free condition also allows for investigations of new chemical systems.

Keywords: Superheavy element chemistry, Gas-filled recoil ion separator, Gas-jet transport system, ^{261}Rf
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INTRODUCTION

Chemistry of superheavy elements (SHEs) with atomic numbers $Z{\geq}104$ has become one of the most exciting and challenging research subjects in nuclear and radiochemistry [1,2]. A most important and interesting question is to clarify chemical properties of these newly synthesized heavy elements and to elucidate the influence of relativistic effects on chemical properties of these heaviest elements [1,3,4]. SHEs are produced in accelerators in heavy-ion-induced nuclear reactions. Extremely low production yields and short half-lives of SHEs force us to conduct rapid and efficient on-line chemical experiments with single atoms. Using gas-jet coupled chemistry apparatuses, chemical properties of SHEs have been studied for elements 104 (Rf) to 108 (Hs) and recently element 112 [1,2,5]. At the same time, many of these successful

experiments have clearly demonstrated the limitations of the applied techniques. Large amounts of background radioactivities from unwanted reaction products become unavoidable for SHEs with higher Z. High-intensity beams from advanced accelerators give rise to a problem in that the plasma formed by the beam in a target chamber significantly reduces the gas-jet transport efficiency. To overcome these limitations, the concept of physical preseparation of SHE atoms has been proposed [1,6]. The pioneering experiments with the recoil transfer chamber (RTC) coupled to the Berkeley Gas-filled Separator (BGS) were very successful [7,8]. The isotope of ^{257}Rf physically separated from the large amount of β-decaying products was identified with a liquid scintillator after a liquid-liquid solvent extraction. However, the very short half-life of ^{257}Rf ($T_{1/2}$=4.7 s) produced in the cold fusion reaction of ^{208}Pb(^{50}Ti,n) imposes stringent time limits on the gas-jet transport as well as the chemical separation [9]. At GSI Helmholtzzentrum für Schwerionenforschung GmbH (GSI), a new gas-filled separator, the TransActinide Separator and Chemistry Apparatus (TASCA), is under commissioning as a preseparator for chemical studies [6,10]. In the recent TASCA commissioning, the fluoride complexation of Rf in a diluted hydrofluoric acid was studied by anion-exchange chromatography using ^{261}Rf ($T_{1/2}$=68 s) produced in the ^{244}Pu(^{22}Ne,$5n$)^{261}Rf reaction [11].

In the RIKEN Linear ACcelerator (RILAC) facility, a prototype gas-jet transport system for the SHE chemistry was installed at the focal plane of the gas-filled recoil ion separator (GARIS) [12]. The performance of the system has been investigated using ^{206}Fr (Z=87), ^{245}Fm (Z=100), and ^{255}No (Z=102) produced in the ^{169}Tm(^{40}Ar,$3n$), ^{208}Pb(^{40}Ar,$3n$), and ^{238}U(^{22}Ne,$5n$) reactions, respectively [12–14]. The results revealed that the GARIS/gas-jet system is a promising tool for next-generation SHE chemistry, i.e., identifying SHE nuclides under low background conditions with high efficiency of the gas-jet transport. Using the GARIS/gas-jet system, we next plan to investigate hot fusion reactions based on a ^{248}Cm target to produce SHE nuclides with long half-lives for chemical experiments. However, very small recoil velocities of evaporation residues (ERs) produced by such asymmetric reactions cause serious problems in the operation of the gas-jet system coupled to the gas-filled separator. The transport efficiency of the separator drastically decreases with decreasing recoil velocity due to the multiple small-angle scattering in the filling gas. A vacuum window foil, which separates the gas-jet chamber from the separator, should be thin enough to allow ERs to pass though and has to withstand a pressure difference of about 1 bar. Since the last experiment with ^{255}No [14], we have developed a new gas-jet chamber having a large focal plane window of 100-mm diameter to efficiently collect ERs. A rotating target system for the use of a radioactive ^{248}Cm material was also installed. A chemistry laboratory was constructed behind the focal plane of GARIS, shielded with 50-cm concrete from the target room. Recently, the most desirable nuclide for Rf chemistry, ^{261}Rf, produced in the very asymmetric ^{248}Cm(^{18}O,$5n$)^{261}Rf reaction was successfully extracted to the chemistry laboratory after the physical separation by GARIS [15]. In this paper, the performance of the GARIS/gas-jet system is demonstrated by referring to the recent ^{261}Rf experiment.

EXPERIMENTAL

A schematic of the experimental setup is shown in Fig. 1. The $^{18}O^{5+}$ ion beam was extracted from RILAC. A $^{248}Cm_2O_3$ target of 280 µg cm^{-2} thickness was prepared by electrodeposition onto a 0.90 mg cm^{-2} Ti backing foil. The eight arc-shaped targets were mounted on a rotating wheel of 100 mm in diameter. The wheel was rotated during the irradiation at 1000 rpm. The beam energy was 95.5 MeV at the middle of the target, and the average beam intensity was 5 particle µA. GARIS was filled with helium at a pressure of 33 Pa. The other details of GARIS are given elsewhere [16]. As shown in Figs. 1a and 1b, the evaporation residues of interest were separated in-flight from beam particles and transfer reaction products with GARIS, and were implanted into the focal plane Si detector (FPD) which consisted of nine Si PIN photodiodes (Hamamatsu S3584). The cycle of the beam-on (100 s) and beam-off (100 s) measurements was performed, because no α peaks of ^{261}Rf were observed in the beam-on spectrum due to large amounts of background events. The α-particle energy resolution of each Si PIN photodiode was about 30 keV FWHM. All events were registered in an event-by-event mode using the VME LIST/PHA module (Iwatsu A3100). The magnetic rigidities of 1.58, 1.73, 1.86, and 2.16 Tm were examined to optimize the transport efficiency of GARIS for ^{261}Rf.
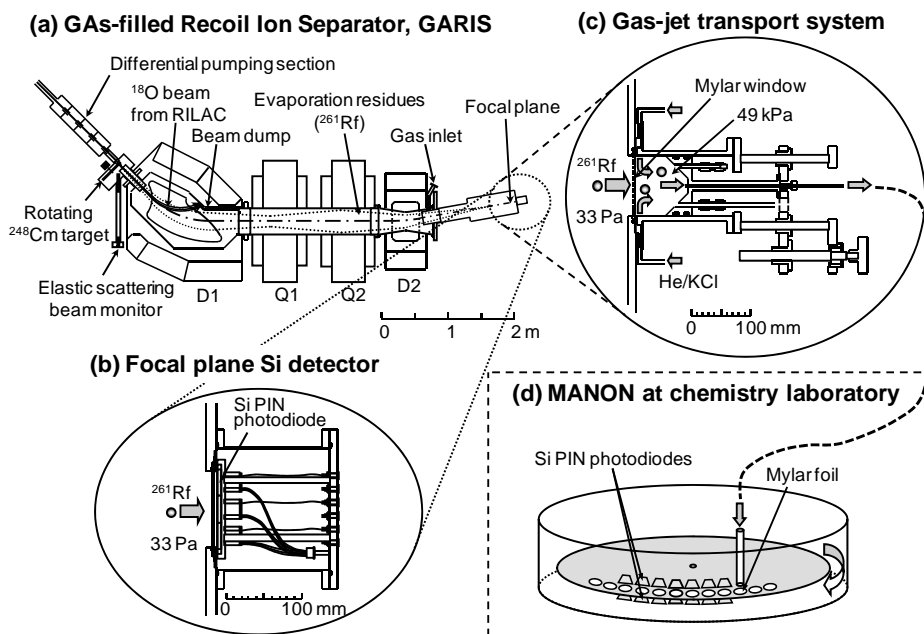

FIGURE 1. A schematic of the experimental setup: (a) RIKEN GAs-filled Recoil Ion Separator GARIS; (b) Focal plane Si detector; (c) Gas-jet transport system; (d) Rotating wheel apparatus MANON for α spectrometry at a chemistry laboratory.

In the gas-jet transport experiments, the reaction products separated with GARIS were guided into the gas-jet chamber of 100-mm i.d.×20-mm depth through a Mylar window of 0.5-mm thickness which was supported by a circular-hole (2.0-mm

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diameter) grid with 78% transparency (see Fig. 1c). The magnetic rigidity was set at 1.73 Tm. In the gas-jet chamber, the [261]Rf atoms were stopped in helium gas, attached to KCl aerosol particles, and were continuously transported through a Teflon capillary (2.0-mm i.d.×10-m length) to the rotating wheel apparatus MANON for α spectrometry (Fig. 1d). The flow rate of the helium gas was 2.0 L min^{-1}, and the inner pressure of the gas-jet chamber was 49 kPa. In MANON, the aerosol particles were deposited on 200-position Mylar foils of 0.5-mm thickness placed at the periphery of a 420-mm diameter stainless steel wheel. The wheel was stepped at 30-s intervals to position the foils between seven pairs of Si PIN photodiodes (Hamamatsu S3204-09). Each detector had an active area of 18×18 mm^2 and a 38% counting efficiency for α particles. The energy resolution was 60 keV FWHM for the detectors which look at the sample from the collection side.

RESULTS AND DISCUSSION

In this work, the magnetic field of GARIS was first optimized for [261]Rf with FPD. In Fig. 2, the transport efficiencies of GARIS for the focal plane of 100-mm diameter are shown as a function of the magnetic rigidity ($B\rho$) of the separator. The cross section for the [248]Cm(^{18}O,5n)[261]Rf reaction, which was measured to be 13 nb at the Japan Atomic Energy Agency tandem accelerator [17], was used to evaluate those transport efficiencies. As shown by a dashed curve in Fig. 2, a least-squares fitting with the Gaussian curve gives a maximum efficiency of 8.0±1.7% at $B\rho$=1.75±0.02 Tm with the resolution of $\Delta B\rho/B\rho$=13±2%.
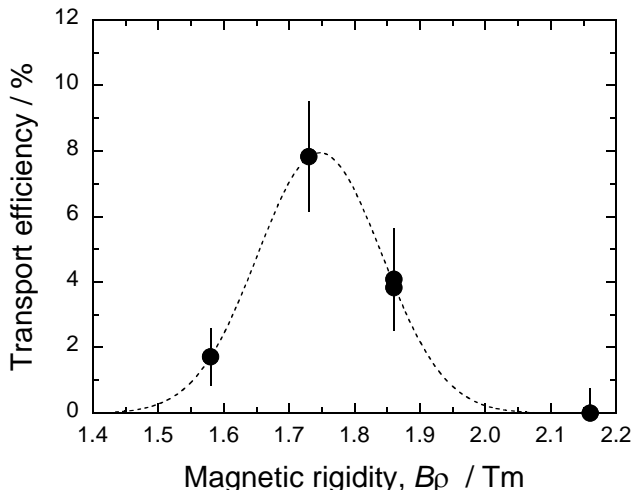

FIGURE 2. Transport efficiency of [261]Rf produced in the [248]Cm(^{18}O,5n)[261]Rf reaction as a function of the magnetic rigidity of GARIS. The dashed curve represents the result of the least-squares fitting with the Gaussian curve with a maximum yield at $B\rho$=1.75±0.02 Tm and a resolution of $\Delta B\rho/B\rho$=13±2%.

Figure 3 shows the sum of α-particle spectra measured in the seven top detectors of MANON. The beam dose of 6.3×10^{17} was accumulated. As shown in Fig. 3, α peaks

of ^{261}Rf (68 s, 8.28 MeV [18]) and its daughter ^{257}No (24.5 s, 8.222 and 8.323 MeV [19]) are clearly seen under the extremely low background conditions. The 7.687-MeV peak is due to ^{214}Po, a descendant of the natural radioisotope ^{222}Rn in the room. The radioactivities due to decays of Po, At, Rn, Fr, Ra, Ac, and Th isotopes, which are largely produced in the transfer reactions on the lead impurity in the target [20], are fully removed by the present system. A total of 168 α events on ^{261}Rf and ^{257}No were registered in the energy range of interest, including 58 time-correlated α pairs. By comparing the spectrum measured with FPD, the gas-jet transport efficiency of ^{261}Rf was evaluated to be 52±12%.
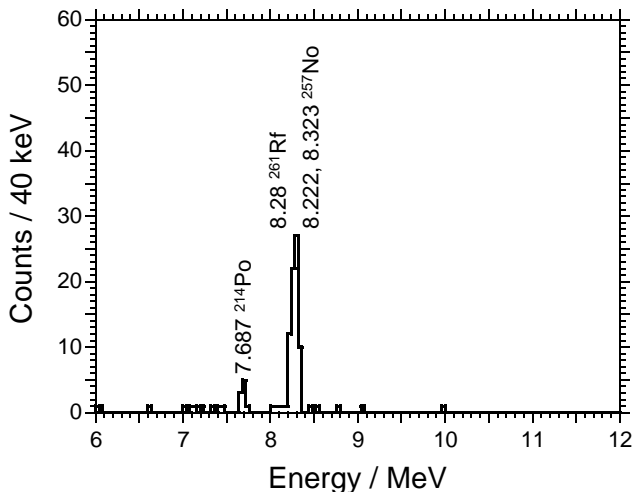

FIGURE 3. Sum of α-particle spectra measured in the seven top detectors of the rotating wheel apparatus MANON for 210 s after the 30-s aerosol collection (Reprinted from [15]). The beam dose of 6.3×10^{17} was accumulated.

SUMMARY AND PERSPECTIVES

In this work, we have successfully produced the isotope of element 104, ^{261}Rf, for chemical studies in the ^{248}Cm-based hot fusion reaction using the gas-jet transport system coupled to GARIS. The α particles of ^{261}Rf were clearly observed with MANON under the desired low background conditions. Under the present experimental condition, the production yield of ^{261}Rf is about 0.5 atoms min^{-1} at the chemistry laboratory. The present result demonstrates that the GARIS/gas-jet system is promising to explore new frontiers in SHE chemistry: (i) the background radioactivities originating from unwanted reaction products are strongly suppressed, (ii) the intense primary heavy-ion beam is absent in the gas-jet chamber, and hence high gas-jet transport efficiency is achieved, and (iii) the beam-free conditions also make it possible to investigate new chemical systems that were not accessible before. In the next phase, we plan to investigate the production and decay properties of heavier SHE nuclides for chemical experiments such as ^{262}Db (Z=105), ^{265}Sg (Z=106), ^{267}Bh (Z=107), and ^{269}Hs (Z=108) based on the ^{248}Cm target. For the future SHE

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chemistry, a gas-chromatograph column directly coupled to GARIS, which enables in-situ complexation and isothermal gas-chromatography of a large variety of volatile compounds of SHEs, is under development. Micro-chemical devices for ion exchange and solvent extraction are also under development together with a gas-jet coupled dissolving unit and an automated α-particle detection system.

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