RIKEN Gas-filled Recoil Ion Separator (GARIS) as a Promising Interface for Superheavy Element Chemistry—Production of Element 104, ^{261}Rf, Using the GARIS/Gas-jet System—

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An isotope of element 104, rutherfordium (^{261}Rf), produced in the ^{248}Cm(^{18}O, 5*n*)^{261}Rf reaction was successfully extracted to a chemistry laboratory using a gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator GARIS. The present system is a promising interface to explore new frontiers in superheavy element chemistry.

Chemical characterization of superheavy elements (SHEs) with atomic numbers $Z \geq 104$ is an extremely interesting and challenging subject in modern 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 experiment with the recoil transfer chamber coupled to the Berkeley gas-filled separator (BGS) was very successful.[7] 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.[8] In the RIKEN linear accelerator (RILAC) facility, the gas-jet transport system for the SHE chemistry was installed at the focal plane of the gas-filled recoil ion separator (GARIS).[9] 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, 3*n*), ^{208}Pb(^{40}Ar, 3*n*), and ^{238}U(^{22}Ne, 5*n*) reactions, respectively.[9,10]

In order to produce SHE nuclides with longer half-lives for chemical experiments, hot fusion reactions based on actinide targets such as ^{244}Pu and ^{248}Cm should be considered. 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 ca. 1 bar. Since the last experiment with ^{255}No, 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. In this work, the most desirable nuclide for Rf chemistry, ^{261}Rf ($T_{1/2} = 68$ s), produced in the very asymmetric ^{248}Cm(^{18}O, 5*n*) reaction was successfully extracted to the chemistry laboratory after the physical separation by GARIS.

A schematic of the experimental setup is shown in Figure 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 magnetic rigidity of GARIS was set at 1.73 Tm. The evaporation residues of interest were separated in-flight from the beam and the majority of the nuclear transfer products by GARIS and then guided into the gas-jet chamber of 100-mm i.d. $\times$ 20-mm depth through a Mylar window of 0.5-μm thickness which was supported by a circular-hole (2.0-mm diameter) grid with 78% transparency. 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. $\times$ 10-m length) to the rotating wheel apparatus MANON for α spectrometry. 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-μm 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 $\times$ 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. All events were registered in an event-by-event mode.

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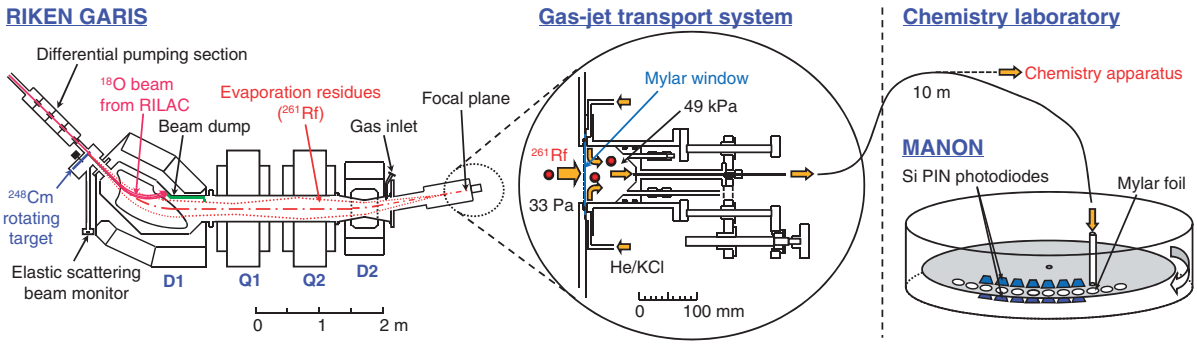

Figure 1. Gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator (GARIS) and the rotating wheel apparatus MANON for α spectrometry placed at the chemistry laboratory.

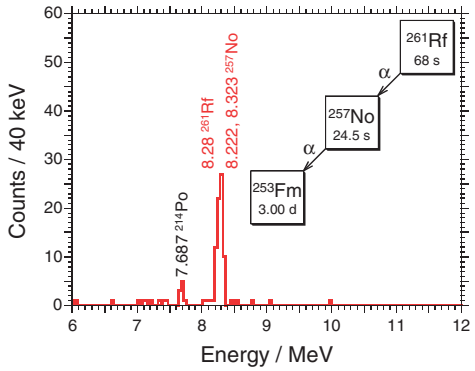

Figure 2. Sum of α-particle spectra measured in the seven top detectors of MANON for 210 s after 30-s aerosol collection.

Figure 2 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 Figure 2, α peaks of ^{261}Rf (68 s, 8.28 MeV)[11] and its daughter ^{257}No (24.5 s, 8.222 and 8.323 MeV)[11] 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,[12] 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 a focal plane Si detector in a separate experiment, the gas-jet transport efficiency of ^{261}Rf was evaluated to be $52 \pm 12\%$. The transport efficiency of GARIS was $7.8 \pm 1.7\%$ for the focal plane of 100-mm diameter, referring to the cross section of 13 nb.[13]

In this work, we have successfully produced ^{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. The production yield of ^{261}Rf at the chemistry laboratory is 0.5 atoms min^{-1} under the present experimental condition. 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.

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