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Development of Gas-jet Transport System Coupled to the RIKEN Gas-filled Recoil Ion Separator GARIS for Superheavy Element Chemistry

H. Haba,*[,a] D. Kaji,[a] H. Kikunaga,[a] T. Akiyama,[a,b] N. Sato,[a,c] K. Morimoto,[a] A. Yoneda,[a] K. Morita,[a] T. Takabe,[d] and A. Shinohara[d]

[a]*Nishina Center for Accelerator Based Science, RIKEN, Wako, Saitama 351-0198, Japan*
[b]*Department of Physics, Saitama University, Sakura, Saitama 338-8570, Japan*
[c]*Department of Physics, Tohoku University, Aoba, Sendai 980-8578, Japan*
[d]*Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan*

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A gas-jet transport system for the superheavy element chemistry was coupled to the gas-filled recoil ion separator GARIS at the RIKEN Linear Accelerator. The performance of the system was investigated using ^{206}Fr and ^{245}Fm produced in the ^{40}Ar-induced reactions on ^{169}Tm and ^{208}Pb targets, respectively. Alpha particles of ^{206}Fr and ^{245}Fm separated with GARIS and transported by the gas-jet were measured with a rotating wheel system for α spectrometry under low background condition. The high gas-jet efficiency of over 80% was found to be independent of the beam intensity up to 2 particle μA. These results suggest that the GARIS/gas-jet system is a promising tool for future superheavy element chemistry at RIKEN.

1. Introduction

The chemistry of the 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] Experimental studies on chemical properties of SHEs have been performed for elements 104 to 108 and very recently element 112.[1,2] The production rates of the SHE nuclides, however, are extremely low, i.e., atoms per minute for elements 104 (Rf) and 105 (Db) down to atoms per hour or day for elements 106 (Sg) to 108 (Hs),[2] and their half-lives are less than ~1 min. This situation forces us to perform rapid and efficient on-line chemical experiments with "single atoms" using heavy-ion accelerators. The gas-jet transport technique has been commonly used to transfer SHE atoms from a target chamber to various chemistry apparatuses. Here, nuclear reaction products recoiling out of a target are stopped in the chamber filled with helium gas, and are often attached to aerosol particles such as potassium chloride (KCl) and carbon. The gas and activity-laden aerosol particles are swept out through a capillary to a distant site where chemistry apparatuses and detectors are equipped.

SHE atoms are produced among large amounts of background radioactivities which hinder the detection of decay signals of SHE nuclides. Recently available high-intensity beams of more than 1 particle μA (pμA) also give rise to a serious problem in that the plasma formed by the beam in the target chamber significantly reduces the gas-jet transport efficiency. To overcome these situations, it has been proposed that a recoil separator for nuclear physics research on SHEs should be coupled to the chemistry system with the aid of the gas-jet transport technique mentioned above.[1,3] With this method, background radioactivities originating from other reaction products are largely removed. The high and stable gas-jet efficiencies are obtained owing to the condition free from plasma. Furthermore, chemical reactions of various compounds can be studied by directly feeding complexing reagents into the gas-jet chamber without aerosol materials. The first experiment with the recoil transfer chamber (RTC) coupled to the Berkeley Gas-filled Separator (BGS) was very successful.[4,5] 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. Thereafter, the BGS/RTC system was used in the model experiments of Rf (References 6–8) and Hs (Reference 9). At Gesellschaft für Schwerionenforschung (GSI), the components of the former HElium Charge-exchange Kaleidoscope (HECK) separator are being used to set up a dedicated separator for the SHE chemistry.[3]

In the RIKEN Linear Accelerator (RILAC) facility, the gas-filled recoil ion separator GARIS has been used to search for the heaviest SHE nuclides.[10–13] The isotopes of ^{271}Ds, ^{272}Rg, and ^{277}112 found at GSI were confirmed with better statistics and with new spectroscopic information,[10–12] and a new isotope of element 113, ^{278}113, was successfully synthesized.[13] GARIS is expected to give us low background condition and high transport efficiencies for SHEs. In this work, we have installed a gas-jet transport system at the focal plane of GARIS to start the SHE chemistry in RIKEN. The performance of the system was appraised using ^{206}Fr and ^{245}Fm produced in the ^{169}Tm(^{40}Ar,3n)^{206}Fr and ^{208}Pb(^{40}Ar,3n)^{245}Fm reactions, respectively.

2. Experimental

A schematic of the experimental setup is shown in Figure 1. The $^{40}Ar^{9+}$ ion beam was extracted from RILAC. The metallic ^{169}Tm and ^{208}Pb targets of 120 and 420 μg cm^{-2} thicknesses, respectively, were prepared by vacuum evaporation on 30 μg cm^{-2} carbon backing foils. Sixteen targets were mounted on a rotating wheel of 30 cm in diameter. The wheel was rotated during the irradiation at 3000 rpm. The beam energies were 169.7 MeV for ^{169}Tm and 198.6 MeV for ^{208}Pb at the middle of the targets. At these incident energies, the cross section for the ^{169}Tm(^{40}Ar,xn)$^{209-x}$Fr reactions (x = 2 + 3) is 376 ± 7 μb,[14] while that for ^{208}Pb(^{40}Ar,3n)^{245}Fm is 15 ± 5 nb.[15] The beam intensity was monitored by measuring elastically scattered projectiles with a Si PIN photodiode (Hamamatsu S1223) mounted at 45° with respect to the beam axis. The typical beam intensity was 2 pμA. GARIS was filled with helium at a pressure of 88 Pa.

*Corresponding author. E-mail: haba@riken.jp. Fax: +81-48-461-5301.

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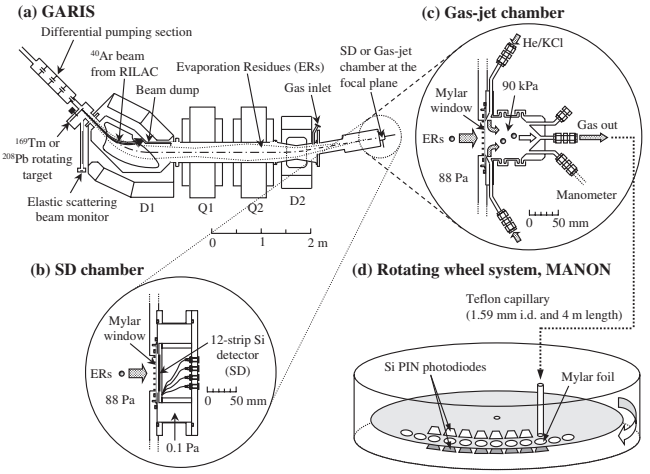

Figure 1. A schematic of the experimental setup: (a) RIKEN gas-filled recoil ion separator GARIS; (b) 12-strip Si detector (SD) chamber; (c) Gas-jet chamber coupled to GARIS; (d) Rotating wheel system MANON for α spectrometry.

The magnetic rigidities were set at 1.64 Tm for ^{206}Fr and at 2.01 Tm for ^{245}Fm. The other details of GARIS are given elsewhere.[11]

As shown in Figures 1(a) and 1(b), the evaporation residues of interest were separated in-flight from beam particles and transfer reaction products with GARIS, and were implanted into a 12-strip Si detector (SD) of 60 × 60 mm^2 (Hamamatsu 12CH PSD) through a Mylar window of 3.5 ± 0.1 μm thickness which was supported with a stainless-steel honeycomb grid with 92.5% transparency and of 60 mm diameter. Alpha-particle measurements of ^{206}Fr were conducted for 2600 s under the beam-on condition and for 300 s after a 30-s beam irradiation. In the ^{245}Fm experiment, the cycle of the beam-on (5 s) and beam-off (15 s) measurements was repeated 1265 times, because no α peak of ^{245}Fm was observed in the beam-on spectrum due to large amounts of background events. The α-particle energy resolution of SD was 50 keV FWHM. All events were registered in an event-by-event mode.

In the gas-jet transport experiments, ^{206}Fr and ^{245}Fm separated by GARIS were guided into the gas-jet chamber of 60 mm length as shown in Figure 1(c). The reaction products were stopped in the helium gas at 90 kPa, attached to KCl aerosol particles generated by sublimation of the KCl powder at 620 °C, and were continuously transported through a Teflon capillary (1.59 mm i.d., 4 m length) to the rotating wheel system MANON for α spectrometry (Figure 1(d)) which was the compact one of the Measurement system for the Alpha-particle and spontaneous fissioN events ON-line developed at Japan Atomic Energy Agency (JAEA).[16] The flow rate of the helium gas was 5 L min^{-1}. In the ^{206}Fr experiment, the temperature of the KCl aerosol generator was varied from 540 to 640 °C in steps of about 20 °C to optimize the gas-jet transport efficiency. In MANON, aerosol particles were deposited on Mylar foils of 0.68 μm thickness and 20 mm diameter placed at the periphery of a 40-position stainless steel wheel of 420 mm diameter. The wheel was stepped at 30-s and 2-s intervals for ^{206}Fr and ^{245}Fm, respectively, 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 α-particle energy resolution was 60 keV FWHM for the detectors which look at the sample from the collection side (top detectors).

3. Results and Discussion

Figure 2(a) shows an α-particle spectrum measured in the ^{206}Fr experiment for 2600 s in the 6th strip (middle) of SD under the beam-on condition. Alpha peaks of ^{206}Fr ($T_{1/2}$ = 15.9 s, E_α = 6.790 MeV)[17] and ^{205}Fr (3.85 s, 6.915 MeV)[17] and of their daughter nuclides ^{202}At (182 s, 6.135 MeV; 184 s, 6.228 MeV)[17] and ^{201}At (89 s, 6.344 MeV)[17] were identified. The broad component above 7 MeV is associated with the implantation of evaporation residues (ERs) and target recoils (TRs). Figure 2(b) shows an α-particle spectrum measured in the 6th strip of SD for 300 s after the 30-s beam irradiation. The decay curve of the 6.790-MeV α peak of ^{206}Fr is shown in the inset of Figure 2(b). The half-life of ^{206}Fr was determined to be 15.3 ± 0.2 s, which is in agreement with the literature value of 15.9 ± 0.2 s (Reference 17) within 2σ. The contribution of the α particles of ^{207}Fr (14.8 s, 6.768 MeV)[17] produced in the ^{169}Tm(^{40}Ar,2n)^{207}Fr reaction to the 6.790-MeV peak of ^{206}Fr is found to be negligible from the absence of the 6.087-MeV peak of its daughter nuclide ^{203}At (7.4 min, 6.087 MeV)[17] in Figure 2(a). On the other hand, the α-particle spectrum measured in the first top detector of MANON for 30 s after the 30-s aerosol collection is compared in Figure 2(c). The α peak of ^{206}Fr is clearly seen in

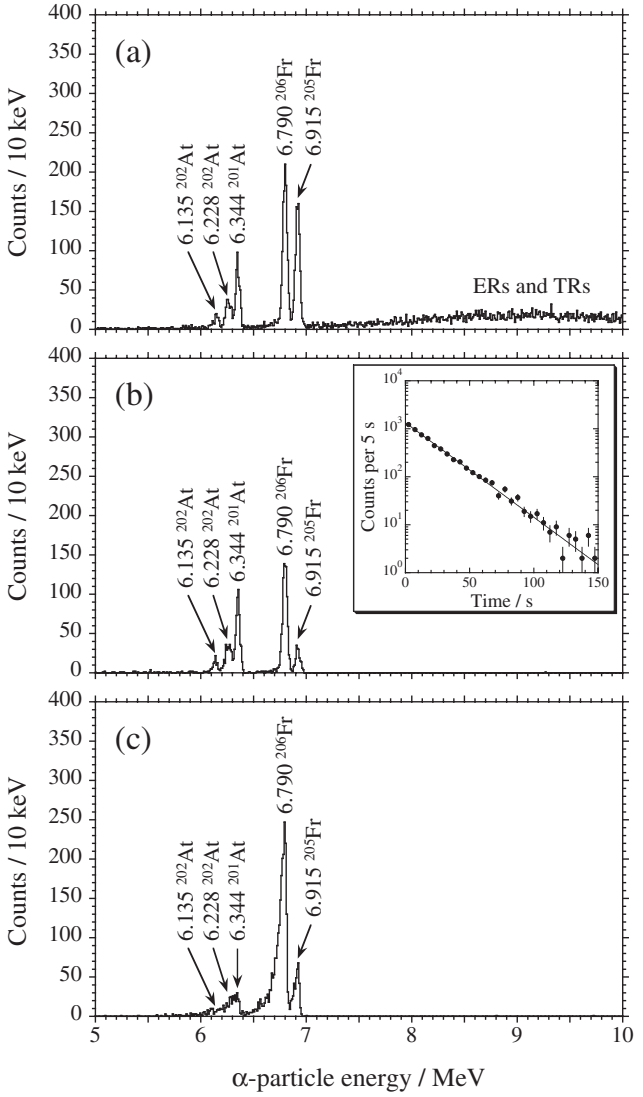

Figure 2. (a) Alpha-particle spectrum measured in the ^{206}Fr experiment for 2600 s in the 6th strip (middle) of the 12-strip Si detector (SD) under the beam-on condition. The ^{40}Ar beam intensity was 0.017 pμA. (b) Alpha-particle spectrum measured in the 6th strip of SD for 300 s after the 30-s beam irradiation. The inset shows a decay curve of the 6.790-MeV α peak of ^{206}Fr measured in all the strips of SD. The beam intensity was 1.9 pμA. (c) Alpha-particle spectrum measured in the first top detector of the rotating wheel system MANON for 30 s after the 30-s aerosol collection. The beam intensity was 1.8 pμA. The helium flow rate was 5 L min^{-1} and the chamber pressure was 90 kPa. The temperature of the KCl aerosol generator was 620 °C.

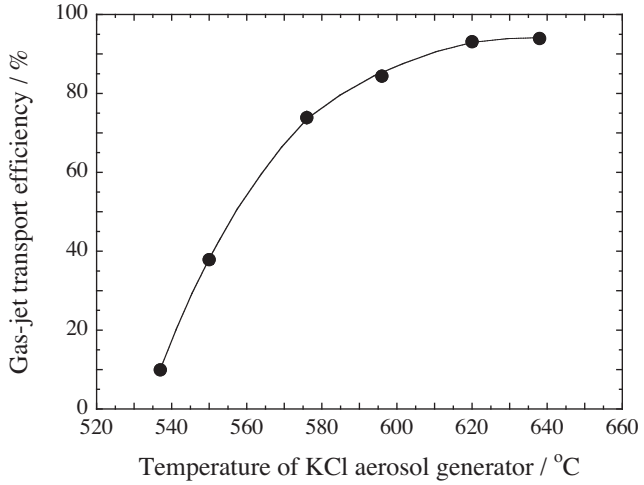

Figure 3. Variation of the gas-jet transport efficiency of ^{206}Fr as a function of the temperature of KCl aerosol generator. The helium flow rate was 5 L min^{-1} and the chamber pressure was 90 kPa. The ^{40}Ar beam intensity was 1.7 pμA.

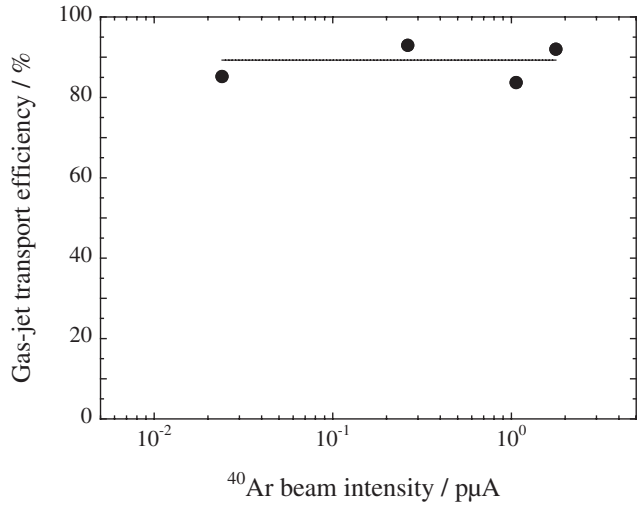

Figure 4. Variation of the gas-jet transport efficiency of ^{206}Fr as a function of the ^{40}Ar beam intensity. The helium flow rate was 5 L min^{-1} and the chamber pressure was 90 kPa. The temperature of the KCl aerosol generator was 620 °C.

the spectrum, indicating that the gas-jet transport of ^{206}Fr to MANON was successfully conducted after the physical separation by GARIS. The transport efficiency of the gas-jet system was evaluated using the 6.790-MeV α peak of ^{206}Fr.

In Figure 3, the gas-jet transport efficiencies of ^{206}Fr are shown as a function of the temperature of the KCl aerosol generator. The efficiency increases smoothly with an increase of the temperature and attain to over 90% at 620 °C. The variation of the gas-jet efficiencies of ^{206}Fr is shown in Figure 4 as a function of the ^{40}Ar beam intensity. The efficiencies are constant irrespectively of the beam intensities up to 2 pμA. In the conventional gas-jet system in that the beam passes through the target chamber, the gas-jet efficiency decreases due to the increasing plasma condition induced by the intense beam. As an example, we previously measured the gas-jet efficiencies of ^{173}W produced in the ^{nat}Gd(^{22}Ne,xn) reaction without the beam separation by GARIS. It was found that the gas-jet efficiency of ^{173}W drastically decreases from 40% at 6.6 pnA to 25% at 0.5 pμA with increasing beam intensity. Since the beam is separated by GARIS in the present experiment, such a decrease of the gas-jet efficiency is not observed for ^{206}Fr as shown in Figure 4.

Alpha-particle spectra of ^{245}Fm under the beam-on and beam-off conditions are shown in Figures 5(a) and 5(b), respectively. Although no α peaks are seen in the beam-on

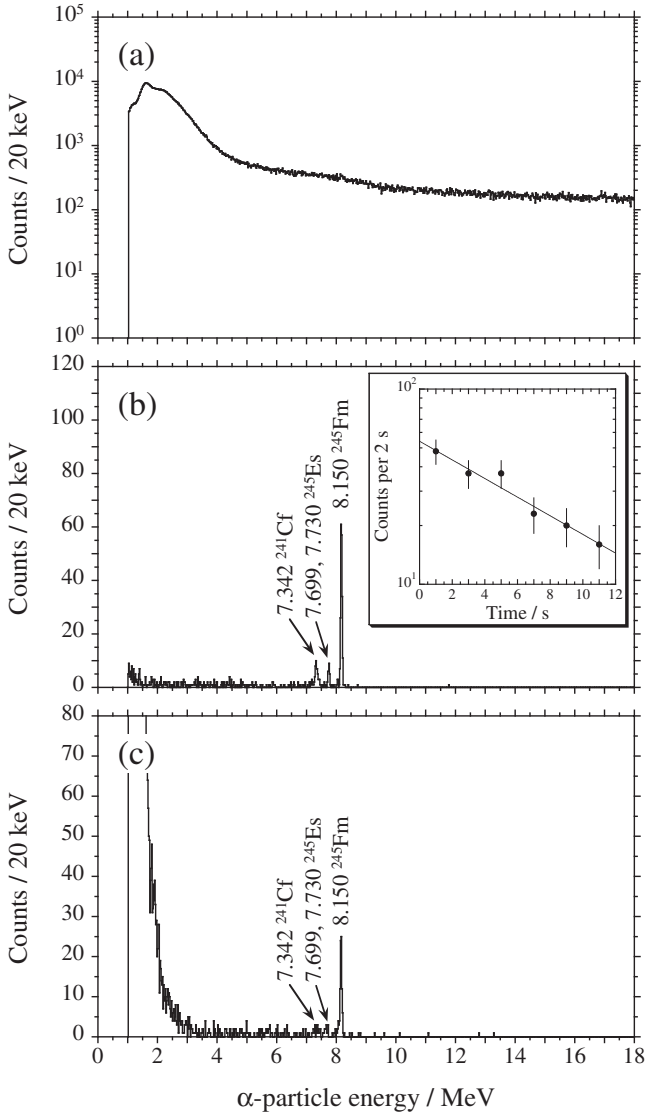

Figure 5. Sum of α-particle spectra measured in the ^{245}Fm experiment in the 12-strip Si detectors (SD) under (a) the beam-on and (b) beam-off conditions. The cycle of the beam-on (5 s) and beam-off (15 s) measurements was repeated 1265 times. The ^{40}Ar beam dose of 6.55×10^{16} was accumulated. The inset shows a decay curve of the 8.150-MeV α peak of ^{245}Fm. (c) Sum of α-particle spectra measured in the seven top detectors of the rotating wheel system MANON for 14 s after the 2-s aerosol collection. The 2-s aerosol collection was repeated 7289 times. The beam dose was 9.76×10^{16}. The helium flow rate was 5 L min^{-1} and the chamber pressure was 90 kPa. The temperature of the KCl aerosol generator was 620 °C.

spectrum (Figure 5(a)), α peaks of ^{245}Fm (4.2 s, 8.15 MeV)[17] and its daughter ^{241}Cf (3.78 min, 7.342 MeV)[17] are clearly seen in the beam-off spectrum (Figure 5(b)). The decay curve of the 8.15-MeV α peak is shown in the inset of Figure 5(b). The half-life of ^{245}Fm was determined to be 6.3 ± 1.3 s, which agrees with the literature value of 4.2 ± 1.3 s.[17] The α peaks around 7.7 MeV would be those of ^{245}Es (1.1 min, 7.699 and 7.730 MeV)[17] produced in the ^{208}Pb(^{40}Ar,p2n)^{245}Es reaction or the EC decay of ^{245}Fm. Background α-particles of, e.g., ^{211}Bi and ^{211m,212m}Po which are produced in the transfer reactions on the ^{208}Pb target[15] are completely removed by GARIS. The separation factors in excess of 10^4 were evaluated for ^{211}Bi and ^{211m,212m}Po based on their cross sections reported in Reference 15.

Compared in Figure 5(c) is the sum of α-particle spectra measured in the seven top detectors of MANON. The 8.15-MeV α peak of ^{245}Fm is clearly seen in the spectrum, and the gas-jet transport efficiency is determined to be 83 ± 9%. Despite of the physical separation by GARIS, one can see a lot of background events in Figure 5(c), especially below 4 MeV.

Since MANON was placed in the target room in this experiment, these background events were caused by large amounts of neutrons and/or γ rays during the irradiation. Very recently, we have constructed a chemistry laboratory isolated with a 50-cm concrete shield from the target room, where the background level is two orders of magnitude lower than that in the target room.

In this work, the high gas-jet efficiencies over 80% were obtained both for ^{206}Fr and ^{245}Fm. The recoil range of ^{206}Fr in helium at 90 k Pa is calculated to be 30 mm based on the SRIM 2006 code,[18] and that of ^{245}Fm is extrapolated to 18 mm from those of the lower Z ions (Z = 68–92) of A = 245. These recoil ranges are short enough as compared with the helium thickness of the gas-jet chamber used in this work (60 mm at 90 kPa). In the conventional gas-jet system in that the beam passes in the recoil chamber, the helium gas is swept out through the capillary outlet to the vertical direction of the beam axis. Therefore, the position of the capillary outlet in the chamber should be adjusted to the recoil ranges of the product nuclei of interest. Since the beam is separated by GARIS in the present system, we can put the capillary outlet at the center end of the chamber (see Figure 1(c)). Thus, helium gas is fed into the chamber through the four inlets directed to the surface of the Mylar window and is swept out thoroughly from the end of the chamber. In addition to the beam-free condition, this smooth helium flow in the chamber would be advantageous to the high gas-jet efficiency.

4. Summary and Perspectives

We have developed the gas-jet transport system coupled to GARIS as a preseparator for the SHE chemistry. The performance of the system was demonstrated using ^{206}Fr and ^{245}Fm produced in the ^{169}Tm(^{40}Ar,3n)^{206}Fr and ^{208}Pb(^{40}Ar,3n)^{245}Fm reactions, respectively. The α particles of ^{206}Fr and ^{245}Fm separated with GARIS and transported by the gas-jet were clearly observed with a rotating wheel system for α spectrometry. The high gas-jet efficiency of over 80% was found to be independent of the beam intensity up to 2 pμA. These results suggest that the GARIS/gas-jet system is a promising tool for future SHE chemistry at RIKEN. Recently, the gas-jet transport of ^{255}No produced in the ^{238}U(^{22}Ne,5n) reaction was also successful with the gas-jet efficiency over 80%. In the future, productions of SHE nuclides with long half-lives for chemical experiments such as ^{261}Rf, ^{262}Db, ^{265}Sg, ^{269}Hs, and ^{283}112 will be studied with the present system using ^{238}U and ^{248}Cm targets.

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