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Startup of superheavy element chemistry at RIKEN

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Abstract. A review is given on the startup of the superheavy element (SHE) chemistry at RIKEN. A gas-jet transport system for the SHE chemistry has been coupled to the gas-filled recoil ion separator GARIS at the RIKEN Linear Accelerator. 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. The α particles of ^{206}Fr and ^{245}Fm separated with GARIS and transported by the gas-jet were identified with a rotating wheel system for α spectrometry under desired low background condition. The high gas-jet efficiencies over 80% were independent of the beam intensities up to 2 particle μA. A gas-jet coupled target system for the production of SHEs was also installed on the beam line of the RIKEN K70 AVF cyclotron. The gas-jet transport of ^{255}No and ^{261}Rf produced in the ^{238}U (^{22}Ne, 5n) ^{255}No and ^{248}Cm (^{18}O, 5n) ^{261}Rf reactions, respectively, was conducted for the future chemical studies of ^{265}Sg via the ^{248}Cm (^{22}Ne, 5n) ^{265}Sg reaction.

PACS. 23.60.+e Alpha decay – 25.70.Gh Compound nucleus – 25.60.Pj Fusion reactions – 25.70.-z Low and intermediate energy heavy-ion reactions

1 Introduction

Recently, the chemistry of superheavy elements (SHEs, atomic numbers $Z \geqslant 104$) has become one of the most exciting and challenging research subjects in nuclear and radiochemistry [1,2]. The chemical properties of newly-discovered and unknown elements, of course, attract a great deal of our interest. Furthermore, influences of the strong relativistic effect on valence electrons of SHEs are often predicted to induce deviations in chemical properties from periodicity based on their lighter homologues in the periodic table [3,4].

The experimental studies on the chemical properties of SHEs have been performed for elements 104 (Rf) to 108 (Hs) and very recently element 112 (E112) [1,2]. The main objectives of these pioneering studies are the placement of the new elements in the periodic table. The results show that elements 104 to 108 are placed onto the expected groups 4 to 8 of the periodic table, respectively. E112 is currently one of the most exciting elements with the question how closely E112 would resemble its lighter homologue Hg [2]. It is also important to study detailed chemical properties of SHEs with high statistics and to compare them with properties deduced from extrapolations of periodicity and from relativistic molecular orbital calculations. The detailed solution chemistry of Rf by an ion-exchange chromatography in HNO_3, HCl, HF, and HF/HNO_3 was very successful at the Japan Atomic Energy Agency (JAEA) Tandem Facility [5].

We are planning to start SHE chemistry at RIKEN using the RIKEN Linear Accelerator (RILAC) and the K70 AVF cyclotron. A gas-jet transport system was coupled to the gas-filled recoil ion separator GARIS as a preseparator for the SHE chemistry. This system is expected to provide new methodologies for the SHE chemistry: identification of SHE nuclides under extremely low background condition, high and stable efficiencies of the gas-jet transport, and direct chemical reactions with a large variety of compounds. A conventional gas-jet coupled target system for the production of SHEs was also installed on the beam line of the AVF cyclotron for the detailed chemistry of SHEs with $Z \geqslant 106$. In this paper, present status and perspectives of the SHE chemistry at RIKEN are summarized.

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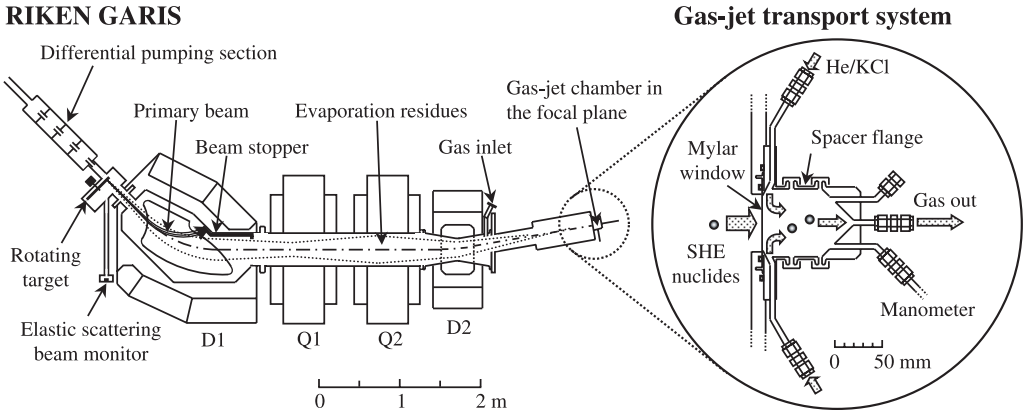

Fig. 1. Schematic of the gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator GARIS at the RIKEN linear accelerator.

2 Development of the GARIS/gas-jet system for SHE chemistry

2.1 Gas-jet transport system coupled to GARIS

The SHE atoms are produced at extremely low production rates among much larger amounts of background activities which hinder the detection of decays of the SHE nuclides of interest. 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 beams significantly decreases gas-jet transport efficiencies. 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 [6]. With this method, background activities that cannot be effectively separated from SHEs, such as those from Po or Rn isotopes, are largely removed. The high and stable gas-jet transport efficiencies are obtained without the plasma condition caused by the beam. 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 [7,8]. The isotope of ^{257}Rf physically separated from the large background caused by β-particles was identified with a liquid scintillator after a liquid-liquid solvent extraction into 0.25 M dibutyl-phosphoric acid in toluene from 6 M HNO_3 with SISAK. Thereafter, the BGS/RTC system has been used in the model experiments of Rf [9–11] and Hs [12]. 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 chemistry experiments [6].

In the RILAC Facility, the gas-filled recoil ion separator GARIS is now in operation to search for the heaviest SHE nuclides such as ^{271}Ds, ^{272}Rg, ^{277}112, and ^{278}113 [13–16]. GARIS gives us extremely low background condition and high transport efficiencies for SHEs. Thus, we have installed a gas-jet transport system in the focal plane of GARIS to start the SHE chemistry at RIKEN. A schematic of the system is shown in Figure 1. The gas-jet chamber at an inner pressure of ∼100 kPa is isolated from GARIS at ∼100 Pa with a very thin Mylar-vacuum window supported with a stainless-steel honeycomb grid with 92.5% transparency and of 60 mm diameter. This grid can support the Mylar foils down to 2.4 μm thickness. As shown in Figure 1, the recoiling SHEs are separated in-flight from the beam and the majority of the nuclear transfer products by GARIS, and are guided into the gas-jet chamber through the Mylar window. The helium gas, often seeded with aerosol particles, is fed into the chamber through the four inlets and is swept out through a Teflon capillary to chemistry apparatuses. The volume of the chamber is variable for ranges of product nuclei of interest with spacer flanges: 70 mm i.d. × 30, 60, and 90 mm long. Recently, the performance of the GARIS/gas-jet 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.

2.2 Production and gas-jet transport of ^{206}Fr and ^{245}Fm

The $^{40}Ar^{9+}$ ion beam was extracted from RILAC. The metallic ^{169}Tm and ^{208}Pb targets of 120 and 420 $\mu g\,cm^{-2}$ thicknesses, respectively, were prepared by vacuum evaporation on a 30 $\mu g\,cm^{-2}$ carbon backing foil. Sixteen targets were mounted on a rotating wheel of 30 cm in diameter. The wheel was rotated during the irradiation at 2000 or 3000 rpm. The beam energies were 170 MeV for ^{169}Tm and 199 MeV for ^{208}Pb at the middle of the target. 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 [17], while that for ^{208}Pb (^{40}Ar, $3n$) ^{245}Fm is 15±5 nb [18]. The beam intensity was monitored by measuring elastically scattered projectiles with a Si PIN photodiode (Hamamatsu S1223) mounted at 45° with respect to the incident beam direction. The typical beam intensity was 2 pμA.

The reaction products of interest were separated with GARIS, and were guided into the 60-mm deep gas-jet chamber through the Mylar window of 3.5 μm thickness. The separator was filled with helium gas at a pressure of 88 Pa. The magnetic rigidities of GARIS were set at 1.64 and 2.01 Tm for ^{206}Fr and ^{245}Fm, respectively. The

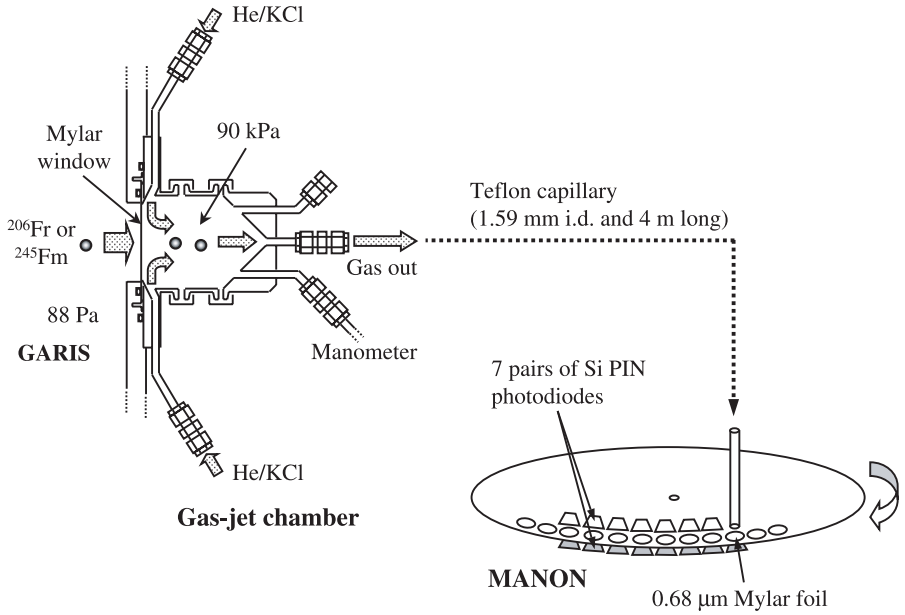

Fig. 2. Schematic of the GARIS/gas-jet system and of the rotating wheel system MANON for α spectrometry.

schematic of the gas-jet transport system is shown in Figure 2. In the gas-jet chamber, the reaction products separated with GARIS were stopped in helium gas, attached to KCl aerosols generated by sublimation of the KCl powder at 620 °C, and continuously transported through a Teflon capillary (1.59 mm i.d., 4 m long) to the rotating wheel system for α spectrometry, which was the compact one of the Measurement system for the Alpha-particle and spontaneous fissioN events ON-line (MANON) developed at JAEA [19]. The helium flow rate was 5 L min^{-1} and the inner pressure of the chamber was 90 kPa. In MANON, the reaction products were deposited on the 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. After the aerosol collection, the wheel was stepped at 30- 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 top detectors. All events were registered in an event-by-event mode.

To evaluate the number of the ^{206}Fr and ^{245}Fm atoms that passed through the Mylar window, the gas-jet chamber was replaced with a detector chamber equipped with a 12-strip Si detector of 60 × 60 mm^2 (Hamamatsu 12CH PSD). The α-particle energy resolution of PSD was 50 keV FWHM.

2.3 Performance of the GARIS/gas-jet system

In Figure 3a, the α-particle spectrum measured for 2600 s in the 6th strip (center) of PSD under beam-on condition is shown. The inset of the figure is an expansion of the energy range of 5.5–7.5 MeV. The α peaks of ^{206}Fr

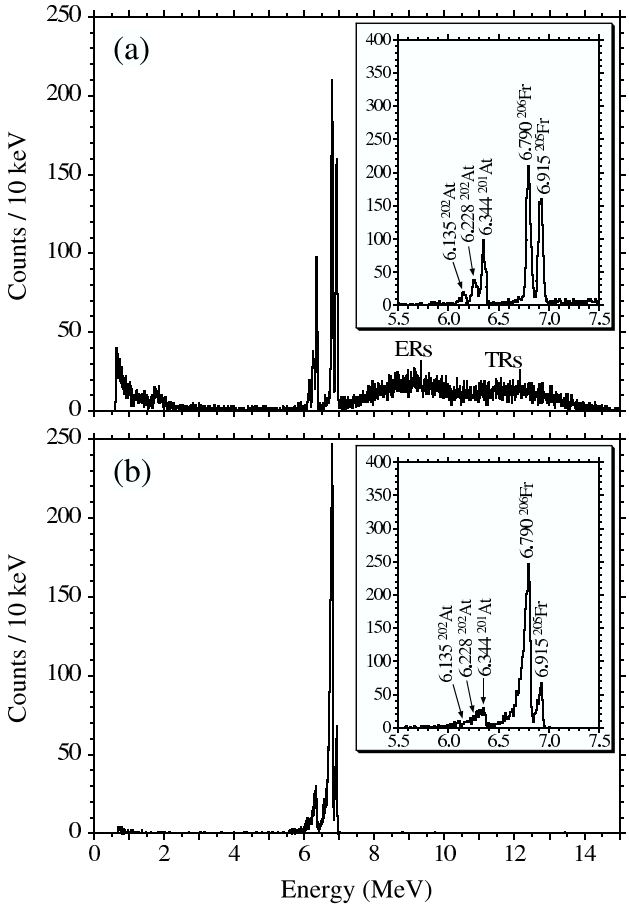

Fig. 3. (a) Alpha-particle spectrum measured for 2600 s in the 6th strip (center) of the 12-strip PSD under the beam-on condition and (b) α-particle spectrum measured for 30 s in the first top detector of the rotating wheel system MANON after the 30-s aerosol collection. The insets of the figures are expansion of the energy range of 5.5–7.5 MeV.

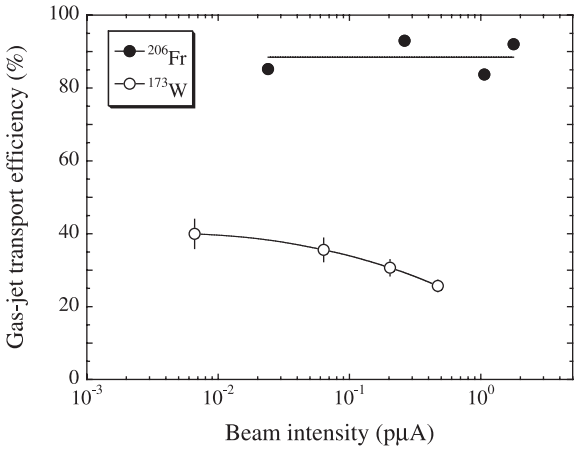

Fig. 4. Variation of the gas-jet transport efficiency of ^{206}Fr (closed circles) and ^{173}W (open circles) as a function of the beam intensity. The isotope ^{206}Fr was produced by the ^{169}Tm (^{40}Ar, $3n$) ^{206}Fr reaction and transported by the gas-jet to the rotating wheel system MANON after the physical separation by GARIS, while ^{173}W was produced by ^{nat}Gd (^{22}Ne, xn) ^{173}W and transported to MANON without the GARIS separation.

($T_{1/2}$ = 15.9 s, E_α = 6.790 MeV [20]) and ^{205}Fr (3.85 s, 6.915 MeV [20]) and of their daughter nuclides ^{202}At (182 s, 6.135 MeV; 184 s, 6.228 MeV [20]) and ^{201}At (89 s, 6.344 MeV [20]) are identified. 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 3b. The broad components above 7 MeV in PSD, which correspond to the implantation of evaporation residues (ERs) and of target recoils (TRs), disappear after the gas-jet transport to MANON as expected (Fig. 3b). The gas-jet transport efficiencies of ^{206}Fr were determined based on the 6.790-MeV peak measured both in PSD and MANON.

In Figure 4, the gas-jet efficiencies of ^{206}Fr are shown by closed circles as a function of the ^{40}Ar beam intensity. The high gas-jet efficiencies over 80% are obtained, and they are independent of the beam intensity. Compared with open circles in Figure 4 are the data of ^{173}W produced in the ^{nat}Gd (^{22}Ne, xn) reaction at the RIKEN K70 AVF cyclotron and transported by the gas-jet without the physical separation by GARIS. Due to the plasma condition induced by the beam in the chamber, the gas-jet efficiencies of ^{173}W decrease from 40% at 6.6 pnA to 25% at 0.5 pμA with an increase of the ^{22}Ne beam intensity. Since the primary beam is separated with GARIS in the ^{169}Tm (^{40}Ar, $3n$) ^{206}Fr experiment, such a decrease is not seen for ^{206}Fr up to 2 pμA studied in this work.

The α-particle spectra of ^{245}Fm measured with PSD and MANON under beam-on condition are shown in Figures 5a and 5b, respectively. The beam doses of 6.55×10^{16} and 9.76×10^{16} were accumulated in the PSD and MANON experiments, respectively. Although large amounts of background events are seen in the PSD spectrum (Fig. 5a), the 8.15 MeV peak of ^{245}Fm is clearly identified in MANON as shown in Figure 5b. The background activities such as Po isotopes, which are produced in the transfer reactions on the ^{208}Pb target, are completely removed by the GARIS/gas-jet system. To evaluate the gas-jet efficiency of ^{245}Fm, the cycle of the beam-on (5 s) and beam-off (15 s) measurements was repeated, because no α peak of ^{245}Fm was identified in the beam-on PSD spectrum (Fig. 5a). Thus, the gas-jet efficiency of ^{245}Fm is determined to be 83 ± 9% based on the 8.15-MeV peak in the beam-off PSD spectrum. The transport efficiency of GARIS is 43 ± 4% by assuming the cross section for the ^{208}Pb (^{40}Ar, $3n$) ^{245}Fm reaction to be 15 nb [18].

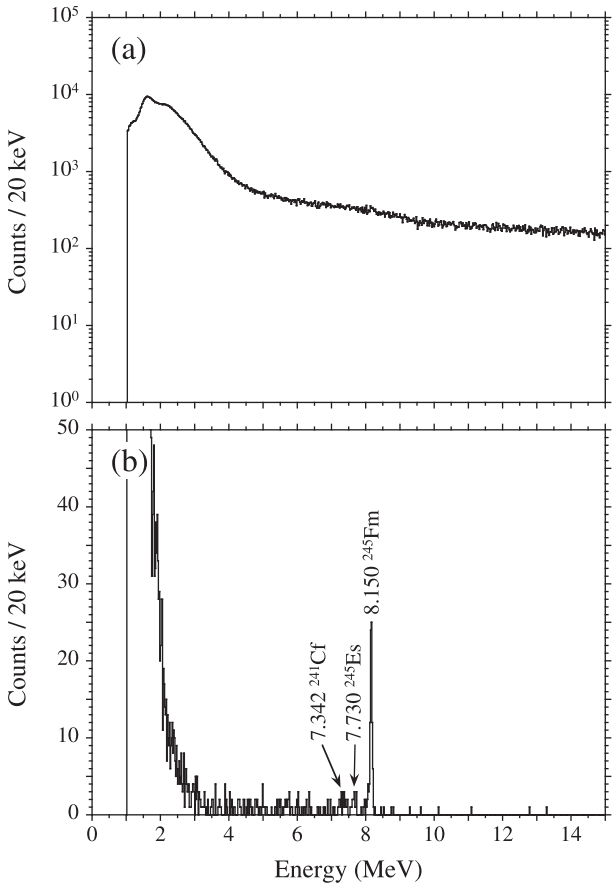

Fig. 5. (a) Sum of α-particle spectra measured in the 12 strip detectors of PSD under the beam-on condition and (b) sum of α-particle spectra measured in the seven top detectors of the rotating wheel system MANON.

Despite of the gas-jet transport after the GARIS separation, one can see some background events in Figure 5b, especially bellow 4 MeV. Since MANON was placed in the target room in this experiment, those background events were mainly due to neutrons and/or γ rays during the irradiation. We are now constructing a chemistry laboratory isolated with a 50-cm concrete shield from GARIS, where the background level is expected to be two orders of magnitude lower than that in the target room.

3 SHE chemistry at the RIKEN K70 AVF cyclotron

3.1 Gas-jet coupled target system for the production of SHEs

As mentioned in introduction, the detailed Rf chemistry has been successfully conducted in HNO_3, HCl, HF, and HF/HNO_3 by the ion-exchange method at JAEA [5]. We intend to extend the series of these experiments to heavier elements than Rf. The ion-exchange experiments with ^{262}Db produced in the ^{248}Cm (^{19}F, 5n) reaction are scheduled at JAEA by developing a new ion-exchange separation apparatus: automated ion-exchange separation apparatus coupled with the Detection system for Alpha spectroscopy II (AIDA II). For heavier elements such as Sg and Hs, we plan to use the RIKEN K70 AVF cyclotron. Recently, we installed a gas-jet coupled target system and a safety system for the usage of radioactive targets on the beam line of the AVF cyclotron. In this work, the performance of the system was investigated using ^{255}No and ^{261}Rf produced in the ^{238}U (^{22}Ne, 5n) ^{255}No and ^{248}Cm (^{18}O, 5n) ^{261}Rf reactions, respectively.

3.2 Production and gas-jet transport of ^{255}No and ^{261}Rf

The $^{22}Ne^{7+}$ and $^{18}O^{6+}$ ion beams were delivered from the RIKEN K70 AVF cyclotron. A ^{238}U target of 630 μg cm^{-2} thickness and a ^{248}Cm target of 480 μg cm^{-2} thickness were prepared by electrodeposition onto a beryllium backing foil of 2.0 mg cm^{-2} thickness. The beams first passed through a beryllium vacuum window (1.9–3.2 mg cm^{-2}), the helium cooling gas (0.09 mg cm^{-2}), the beryllium target backing, and finally entered the target material. The beam energies on target were 105.9, 107.3, 109.0, 113.4, 116.6, and 120.9 MeV for ^{22}Ne and 94.4 MeV for ^{18}O. The beam intensity was approximately 350 pnA. Reaction products recoiling out of the target were stopped in helium gas at 130 kPa (98 kPa)[1], attached to KCl aerosols generated at 640 °C (620 °C)[1], and transported through a Teflon capillary of 2.0 mm i.d. and 45 m long (11 m long)[1] to the rotating wheel system MANON for α spectrometry (see Fig. 2). The flow rate of the helium gas was 2.0 L min^{-1} (5.0 L min^{-1})[1].

In the α spectrum, the α particles of ^{255}No ($T_{1/2}$ = 3.1 min, E_α = 7.620−8.312 MeV [20]) were identified and its radioactivity was evaluated by the two-component decay curve analysis of ^{255}No and ^{254}No (55 s, 8.093 MeV [20]). The gas-jet efficiency was determined to be 50% from radioactivities of the daughter nuclide ^{255}Fm by the catcher foil method with the aid of chemical separation. It was found that the maximum cross section of the ^{238}U (^{22}Ne, 5n) ^{255}No reaction is 90 nb at 113 MeV, though the excitation function measured by Donets et al. [21] shows the peak of 200 nb at 118 MeV. It is noted here that the measurements by Donets et al. [21] were made based on the α decay of the daughter nuclide ^{251}Fm only in ∼1%. The peak cross section corrected for the latest α-decay branch of 1.80% [20] (∼100 nb) is good agreement with the present result.

[1] The values given in parenthesis refer to the condition for the ^{248}Cm (^{18}O, 5n) ^{261}Rf experiment.

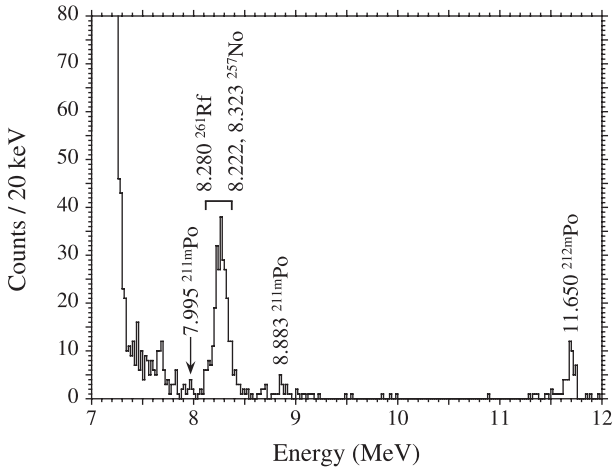

Fig. 6. Sum of α-particle spectra measured in the seven top detectors of the rotating wheel system MANON for 5 to 210 s after the 30-s aerosol collection.

In the ^{261}Rf experiment, the beam dose of 3.49×10^{16} was accumulated. Figure 6 shows the sum of the α-particle spectra measured in the seven top detectors of MANON for 5 to 210 s after the 30-s aerosol collection. A total of 155 time-correlated α pairs of ^{261}Rf (68 s, 8.28 MeV [20]) and its daughter ^{257}No (25 s, 8.222 and 8.323 MeV [22]) were registered in the α-energy range of interest. The gas-jet efficiency of ^{261}Rf in the present system was evaluated to be 74%, by referring to the cross section of 13 nb at 94 MeV [19].

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 identified with a rotating wheel system for α spectrometry. The high gas-jet efficiencies over 80% are independent of the beam intensity up to 2 pμA as expected. These results suggest that the GARIS/gas-jet system is promising tool for the future SHE chemistry. Very recently, the gas-jet transport of ^{255}No produced in the ^{238}U (^{22}Ne, 5n) reaction was also successful with the gas-jet efficiencies over 80%, though the GARIS transport efficiency was relatively small (about 5% for the focal plane of 60 mm diameter). 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 GARIS/gas-jet system based on the ^{238}U and ^{248}Cm targets. A gas-chromatograph column directly coupled to GARIS, which

would enable isothermal gas-chromatography of a large variety of volatile compounds of SHEs, is under development.

The conventional gas-jet coupled target system for the production of SHEs was also installed on the beam line of the RIKEN K70 AVF cyclotron. The gas-jet transport of ^{255}No and ^{261}Rf produced in the ^{238}U (^{22}Ne, $5n$) ^{255}No and ^{248}Cm (^{18}O, $5n$) ^{261}Rf reactions, respectively, was conducted for the future chemical studies of ^{265}Sg. The experiments to measure the excitation function of the ^{248}Cm (^{22}Ne, $5n$) ^{265}Sg reaction are scheduled. The next-generation chemistry devices such as a microchip for solvent extraction [23] and a flow electrolytic column for electrochemistry [24] are under development in collaboration with Osaka University and JAEA.

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