Coprecipitation experiment with Sm hydroxide using a multitracer produced by nuclear spallation reaction: A tool for chemical studies with superheavy elements†

Y. Kasamatsu,[*1] T. Yokokita,[*1] K. Toyomura,[*1] Y. Shigekawa,[*1] H. Haba,[*2] J. Kanaya,[*2] M. Huang,[*2] Y. Ezaki,[*2] T. Yoshimura,[*3] K. Morita,[*2] and A. Shinohara[*1]

The chemical properties of superheavy elements (SHEs) are expected to deviate from the periodicity of their lighter homologues in the periodic table because of the strong relativistic effects on the valence electronic shells of the heavy atoms. However, it is difficult to perform chemical experiments of SHEs because these nuclides are short-lived and their production rates by nuclear reactions are quite low. These conditions create the need for rapid chemical experiments on a one-atom-at-a-time basis. In addition, for unambiguous identifications of these elements existing as single atoms, it is necessary to detect α particles with characteristic energies. These experimental difficulties limit the variety of chemical investigations of SHEs. So far, most of the methodologies in aqueous chemistry were limited to the ion-exchange method or solvent extraction. The purpose of the present study is to establish a coprecipitation method as a new methodology to explore the chemistry of SHEs. In the present study, a multitracer which contains various radioisotopes (various homologues of SHEs) was used to investigate the applicability of the coprecipitation experiment with Sm hydroxide to SHE chemistry.

The multitracer was produced by irradiating ^{nat}Ta targets (2-μm thickness × 30 foils) with $^{14}N^{7+}$ ions accelerated to 135 MeV per nucleon by the RIKEN ring cyclotron (~1.6 electric μA) for 40 h. Various nuclides, namely various elements, with $Z \leq 73$ (Ta) were produced by nuclear spallation reactions. Only the nuclear reaction products recoiling out of the target foils were attached to KCl aerosols seeded in a He gas and transported from the nuclear reaction chamber to the chemical laboratory by a He gas jet through a PTFE capillary tube.[1)] The transported products were collected on glass filter paper, and were dissolved in 1 mL of 0.01 M HCl.

In a polypropylene (PP) beaker, 20 μL of Sm standard solution (1000 mg L^{-1}, 1 M HNO_3) was added into 220 μL of the prepared multitracer solution. The solution was stirred, and then, 2 mL of the basic solution (dilute and concentrated aqueous NH_3, and 0.10, 1.0, 6.0, and 12 M NaOH solutions) was added. The solution was stirred for 10 s or 10 min at room temperature to determine the chemical reaction time required to reach equilibrium. Then, the solution containing the precipitate was filtrated by suction with a PP membrane filter (eichrom, Resolve® Filters 0.1 μm). Both the precipitate and filtrate were dried using a heater at 100 ºC. The precipitate, filtrate, and PP beaker used were subjected to γ-ray spectrometry with Ge semiconductor detectors. The standard activities of the radiotracers were also determined. The precipitation yields were evaluated from these radioactivities.

The product nuclides were identified based on their characteristic γ-ray energies and corresponding half-lives. In total, we identified 34 elements containing more than 60 nuclides as follows: ^{24}Na, ^{42}K, ^{82m}Rb, ^{127,129}Cs {group 1}, ^{28}Mg, ^{47}Ca, $^{128,(131)}$Ba {group 2}, ^{44,47,48}Sc, $^{(86),87g,(m)}$Y {group 3}, (^{132}La), ^{132,135}Ce, $^{145,146,(147)}$Eu, ^{146,147,149}Gd, $^{149\text{-}153,155}$Tb, ^{152,155,157}Dy, (^{160m}Ho), ^{161}Er, $^{165,(166),167}$Tm, $^{166,(169)}$Yb, $^{169,(170),171,172}$Lu {group 3 (lanthanides)}, $^{(86,87)89}$Zr, ^{170,173}Hf {group 4}, ^{90}Nb, ^{176}Ta {group 5}, ^{93m}Mo {group 6}, ^{96}Tc {group 7}, $^{99m,100,(101m)}$Rh {group 9}, ^{65}Zn {group 12}, ^{67}Ga, ^{110,111}In {group 13}, ^{71,72}As, ^{118m,120m}Sb {group 15}, ^{73}Se, and ^{119m}Te {group 16}. The nuclides listed in parentheses are ones for which the coprecipitation yields could not be determined. Various elements belonging to various groups in the periodic table were included in the multitracer used. The presently prepared multitracer contains the homologues of various (nine) SHEs (group 4–18), and thus, this study is beneficial for the model experiment toward the chemical study on the SHEs.

The coprecipitation yields of radionuclides of groups 1–7, 9, 12, 13, 15, 16 elements with Sm hydroxide and their dependence on the kind and concentration of the prepared basic solutions qualitatively reflect the hydroxide precipitation properties (formation of ammine and hydroxide complexes) of the elements in macro amounts. This finding suggests that the coprecipitation behavior of an element whose chemical properties are unknown can be investigated using the present method, and we can discuss its general hydroxide precipitation properties based on the results obtained for coprecipitation. In addition, the chemical reactions of the majority of the elements in the coprecipitation are sufficiently fast to reach equilibrium within 10 s of stirring although for Mo and Se, relatively slow chemical reactions were observed. Based on the results in the present experiment, we can say that the present coprecipitation method with Sm hydroxide is applicable to SHE chemistry. This methodology will open new chemistry routes (various precipitates) for SHEs.

Reference

1) H. Haba et al.: Radiochim. Acta **93**, 539 (2005).

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*1 Graduate School of Science, Osaka University
*2 RIKEN Nishina Center
*3 Radioisotope Center, Osaka University