Accelerator Applications Research Division
RI Application Research Group
Nuclear Chemistry Research Team

1. Abstract

The Nuclear Chemistry Research Team develops production technologies of unique radioisotopes (RIs) at RIKEN RI Beam Factory (RIBF) and applies them in the research fields of physics, chemistry, biology, engineering, medicine, pharmaceutical and environmental sciences. The purified RIs such as ^{65}Zn, ^{67}Cu, ^{85}Sr, ^{88}Y, and ^{109}Cd are delivered to universities and institutes through Japan Radioisotope Association. We also develop new technologies of mass spectrometry for the trace-element analyses using accelerator techniques and apply them to the research fields such as cosmochemistry, environmental science, archaeology, and so on. We perform various isotopic analyses on the elements such as S, Pd, and Pb using ICP-MS, TIMS, and IRMS. We also develop chemical materials such as metallic ^{238}U, $^{238}UO_2$, and ^{48}CaO for ECR ion sources of the heavy-ion accelerators at RIBF.

2. Major Research Subjects

(1) Research and development of RI production technologies at RIBF
(2) RI application researches
(3) Development of trace element analyses using accelerator techniques and their applications to geoscience and archaeological research fields
(4) Development of chemical materials for ECR ion sources of the heavy-ion accelerators at RIBF

3. Summary of Research Activity

(1) Research and development of RI production technologies at RIBF and RI application researches

Due to its high sensitivity, the radioactive tracer technique has been successfully applied for investigations of the behavior of elements in the fields of chemistry, biology, engineering, medicine, pharmaceutical and environmental sciences. We have been developing production technologies of useful radioisotopes (RIs) at RIBF and conducting their application studies in collaboration with many researchers in various fields. With 30-MeV proton, 24-MeV deuteron, and 50-MeV alpha beams from the AVF cyclotron, we presently produce about 100 RIs from ^{7}Be to ^{211}At. Among them, ^{65}Zn, ^{67}Cu, ^{85}Sr, ^{88}Y, and ^{109}Cd are delivered to Japan Radioisotope Association for fee-based distribution to the general public in Japan. Our RIs are also distributed to researchers under the Supply Platform of Short-lived Radioisotopes for Fundamental Research, supported by MEXT KAKENHI in FY2016–2021. On the other hand, RIs of a large number of elements are simultaneously produced from metallic targets such as ^{nat}Ti, ^{nat}Ag, ^{nat}Hf, ^{197}Au, and ^{232}Th irradiated with a 135-MeV nucl.$^{-1}$ ^{14}N beam from the RIKEN Ring Cyclotron. These multitracers are also supplied to universities and institutes as collaborative researches.

In 2020, we developed production technologies of RIs such as ^{7}Be, ^{28}Mg, ^{43}K, ^{44m}Sc, ^{44}Ti, ^{48}Cr, ^{186}Re, ^{211}At, ^{212}Pb, ^{224}Ra, ^{225}Ac, and ^{229}Pa which were strongly demanded but lack supply sources in Japan. We also investigated the excitation functions for the ^{nat}V(d, x), ^{nat}Gd(d, x), ^{141}Pr(d, x), and ^{nat}Nd(α, x) reactions to quantitatively produce useful RIs. We used radiotracers of ^{28}Mg, ^{211}At, ^{212}Pb, ^{224}Ra, and ^{229}Pa for application studies in chemistry, ^{44m}Sc, ^{67}Cu, ^{186}Re, ^{211}At, and ^{225}Ac in nuclear medicine, and ^{43}K, ^{48}Cr, ^{186}Re, and ^{211}At in engineering. We also produced ^{65}Zn, ^{85}Sr, ^{88}Y, and ^{109}Cd for our scientific researches on a regular schedule and supplied the surpluses through Japan Radioisotope Association to the general public. In 2020, we accepted 6 orders of ^{65}Zn with a total activity of 33 MBq, 5 orders of ^{85}Sr with 9.7 MBq, 1 order of ^{88}Y with 1 MBq, and 1 order of ^{109}Cd with 10 MBq. We also distributed ^{7}Be (0.45 MBq × 1), ^{88}Zr (1 MBq × 2), ^{95}Nb (1 MBq × 1 and 2 MBq × 1), ^{121m}Te (2 MBq × 1), ^{175}Hf (1 MBq × 2), and ^{211}At (5 MBq × 6, 10 MBq × 1, 18 MBq × 2, 50 MBq × 1, 80 MBq × 1, and 100 MBq × 16) under the Supply Platform of Short-lived Radioisotopes for Fundamental Research.

(2) Superheavy element chemistry

Chemical characterization of newly-discovered superheavy elements (SHEs, atomic number $Z \geq 104$) is an extremely interesting and challenging subject in modern nuclear and radiochemistry. We are developing SHE production systems as well as rapid single-atom chemistry apparatuses at RIBF. Using heavy-ion beams from RILAC and AVF, ^{261}Rf ($Z = 104$), ^{262}Db ($Z = 105$), ^{265}Sg ($Z = 106$), and ^{266}Bh ($Z = 107$) are produced in the ^{248}Cm(^{18}O, $5n$)^{261}Rf, ^{248}Cm(^{19}F, $5n$)^{262}Db, ^{248}Cm(^{22}Ne, $5n$)^{265}Sg, and ^{248}Cm(^{23}Na, $5n$)^{266}Bh reactions, respectively, and their chemical properties are investigated.

We installed a gas-jet transport system to the focal plane of the gas-filled recoil ion separator GARIS at RILAC. This system is a promising approach for exploring new frontiers in SHE chemistry: the background radiations from unwanted products are strongly suppressed, the intense primary heavy-ion beam is absent in the gas-jet chamber, and hence the high gas-jet extraction yield is attained. Furthermore, the beam-free condition makes it possible to investigate new chemical systems. To realize aqueous chemistry studies of Sg and Bh, we have been developing a continuous and rapid solvent extraction apparatus which consists of a continuous dissolution apparatus Membrane DeGasser (MDG), a Flow Solvent Extractor (FSE), and a liquid scintillation detector for α/SF-spectrometry. On the other hand, we produced radiotracers of $^{88, 89m}$Zr, ^{95}Nb, ^{175}Hf, and $^{177, 179}$Ta at the AVF cyclotron and conducted model experiments for aqueous chemistry studies on Rf and Db. We also developed a cryogenic RF-carpet gas cell, which will be placed on the focal plane of GARIS and connected to a gas chromatographic apparatus, for the future gas-phase chemistry of the short-lived SHEs (<3 s).

(3) Development of trace element analyses using accelerator techniques and their applications to geoscience and archaeological research fields

We have been developing the ECR Ion Source Mass Spectrometer (ECRIS-MS) for trace element analyses. We renovated the detection system of ECRIS-MS and evaluated its sensitivity and mass resolution power. We equipped a laser-ablation system with an ion source and a pre-concentration system to achieve high-resolution analyses for noble gases such as Kr and Xe.

Using the conventional ICP-MS, TIMS, IRMS, and so on, we studied Pb and S isotope ratios on cinnabar and asphalt samples from ancient ruins in Japan to elucidate the distribution of goods in the archaic society and to reveal the establishment of the Yamato dynasty in the period from Jomon to Tumulus. We established a sampling technique for pigment without any damages on the artifacts or wall paintings, using a S-free adhesive tape. Then, we applied the technique to the analyses of the pigment from Roman ruins (Avinyó in Barcelona, Spain). We also applied the technique to the analyses of the red-color substances on the artifacts such as Kyoden remains (Izumo-city, Shimane prefecture), Renpeijou-ato (Zentsuji-City, Kagawa prefecture) and so on. We also established the method to identify the source mine of vermilion, using sulfur, mercury and lead isotopic analyses, and we applied this method to investigation of vermilion from three representative tombs (Kofunperiod in Japan)

(4) Development of chemical materials for ECR ion sources of the heavy-ion accelerators at RIBF

In 2020, we prepared $^{238}UO_2$ on a regular schedule for ^{238}U-ion accelerations with the 28-GHz ECR of RILAC II.

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List of Publications & Presentations

Publications

[Original Papers]

A. R. Usman, M. U. Khandaker, H. Haba, N. Otuka, and M. Murakami, "Production cross sections of thulium radioisotopes for alpha-particle induced reactions on holmium," Nucl. Instrum. Methods Phys. Res. B **469**, 42 (2020).

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Y. Ohshima, H. Suzuki, H. Hanaoka, I. Sasaki, S. Watanabe, H. Haba, Y. Arano, Y. Tsushima, and N. S. Ishioka, "Preclinical evaluation of new α-radionuclide therapy targeting LAT1: 2-[^{211}At]astato-α-methyl-L-phenylalanine in tumor-bearing model," Nucl. Med. Biol. **90–91**, 15 (2020).

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M. Saito, M. Aikawa, T. Murata, Y. Komori, H. Haba, S. Takács, F. Ditorói, and Z. Szűcs, "Production of ^{169}Yb by the proton-induced reaction on ^{169}Tm," Proc. 2019 Symp. Nucl. Data, JAEA-Conf 2020-001, 79 (2020).

Presentations

[International Conferences/Workshops]

H. Takashima (poster), Y. Koga, K. Onuki, S. Manabe, R. Tsumura, T. Anzai, N. Iwata, M. Yasunaga, W. Yang, T. Yokokita, Y. Komori, D. Mori, H. Haba, H. Fujii, and Y. Matsumura, "Preclinical evaluation of astatine-211-conjugated anti-tissue factor antibody," American Association for Cancer Research (AACR) Virtual Annual Meeting II, online, June 22–24, 2020.

K. Ooe (poster), T. Watabe, Y. Shirakami, D. Mori, T. Yokokita, Y. Komori, H. Haba, and J. Hatazawa, "Production and separation of theranostic radionuclide Ag-111 from Pd target," Society of Nuclear Medicine and Molecular Imaging (SNMMI) Annual Meeting 2020, online, July 11–14, 2020.

H. Haba (invited), "Production and applications of radioisotopes at RIKEN RI Beam Factory—Search for new elements through diagnosis and therapy of cancer—," Symposium on Nuclear Data 2020, Wako, Japan, November 26–27, 2020.

Y. Komori (poster), H. Haba, M. Aikawa, M. Saito, S. Takács, and F Ditrói, "Production cross sections of ^{175}Hf in the ^{nat}Lu(p,xn) and ^{nat}Lu(d,xn) reactions," Symposium on Nuclear Data 2020, Wako, Japan, November 26–27, 2020.

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[Domestic Conferences/Workshops]

高島大輝 (ポスター発表), 古賀宣勝, 大貫和信, 津村遼, 岩田望, 眞鍋史乃, 羽場宏光, 藤井博史, 安永正浩, 松村保広, 「アルファ線放出核種アスタチン—211 結合抗組織因子抗体の開発」, 第 36 回日本 DDS 学会学術集会, 神戸, 2020 年 8 月 28–29 日.

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