

English Abstracts

1S01 Decommissioning of Fukushima Daiichi NPP with Radiochemistry

YAMANA, H.

(Nuclear Damage Compensation and Decommissioning Facilitation Corporation)

For the decommissioning of Fukushima Daiichi NPP (1F), which suffered from severe accident in 2011, various measures have been taken for the unprecedentedly difficult challenges, such as contaminated water issue, recovery of fuel-debris and spent nuclear fuels, radioactive waste management, etc. In this report, the latest status of 1F decommissioning will be given with a special focus on the importance and points of radiochemical subjects.

In the three units of 1F, essential boundaries (fuel cladding, PCV and RPV) were lost, and as a result, some of nuclear fuel materials and fission products are existing in an extremely abnormal condition. They are fuel-debris in reactor building, spent nuclear fuels stored in storage pools, massive amount of rubbles contaminated with leaked radioactive nuclides, radioactive contaminated water, secondary wastes generated from water purification, etc. Thanks to the emergency measures taken during these 9 years, stability has been ensured, and it is expected to start mid and long-term operation to lower with specially devised methods the existing radiological risks by dissolving abnormal situations.

Lot of difficulty is expected in future, in terms of severe radiation conditions, insufficient understanding on the property and behavior of the contaminated substances, and massive amount and diversity of the contaminated substances. The “uncertainty” is the substantial key of this challenge, in which utilization of remote devices and other special technologies, and, it will need thorough and careful preparation and operation to get over this big “uncertainty”. Reduction of the uncertainty will surely improve the safety and cost, but there still exist many problems to be solved, particularly of the insufficient understandings on the radiochemical properties and behaviors of the damaged facilities

and the site. The operation will be greatly improved by radiochemical affairs, such as the enhanced elucidation of the properties and chemical behavior of the wastes and fuel-debris, strengthened capability in analysis and chemical design for decontamination and stabilization, as well as for the contaminated water management, and strengthened evaluation of radiological safety and environmental behaviors.

The importance of the radiochemical approach is common for the clean-up of the world nuclear-legacy sites, which are the results of improper operations in the past. In this paper, the author wants to focus on the importance of sharing among the members of JNRS the meaning and direction of radiochemical researches for the response to the nuclear-legacies.

1A01 Creation of chemical separation methods and the determination of excitation functions for Np-spike production

NAKAJIMA, A.¹, SAKAGUCHI, A.¹, HAYAKAWA, Y.², HABA, H.³, MATSUMURA, N.², TERANISHI, K.², MORITA, R.², YOKOKITA, T.³, KOMORI, Y.³, YANG, W.³, MORI, D.³, HARIN, K.⁴, YAMASAKI, S.¹, ZHENG, J.⁵, SUEKI, K.⁵, YOKOYAMA, A.²
(¹Univ. of Tsukuba., ²Kanazawa Univ., ³RIKEN., ⁴Vienna Univ., ⁵QST)

²³⁷Np ($T_{1/2}=2.14 \times 10^6$ y), which is released into the environment through human nuclear activity and remains in the environment as a harmful mobile nuclide for long periods of time, is also a useful environmental dynamics tracer. However, no comprehensive research has been conducted due to the unavailability of an Np ‘spike’ that can be used to correct chemical yields and determine instrument efficiency. In our search for a new Np-spike we focused on ^{236g}Np ($T_{1/2}=1.54 \times 10^5$ y), and attempted to produce such spike through ²³²Th(⁷Li, xn)^{239-x}Np reactions. With this as our stated purpose, we conducted experiments wherein we irradiated a ⁷Li²⁺ beam onto ²³²Th. We also created appropriate chemical separation methods to extract and purify the Np isotopes

from the irradiated Th. We successfully created chemical separation methods for these experiments using TEVA, TK400 and UTEVA resins. The generation of ^{236m}Np was confirmed in stacked Th foils, and the cross sections of $^{232}\text{Th}(^7\text{Li}, 3n)^{236m}\text{Np}$ for some Li energies were obtained through experiments. The ^{236g}Np and ^{237}Np ratios and concentrations will be given using AMS and SF-ICP MS.

1A02 An effect of incomplete fusion on the Np production in the $^{232}\text{Th} + ^7\text{Li}$ reaction

TERANISHI, K.¹, MORITA, R.¹, HAYAKAWA, Y.¹, SAKAGUCHI, A.², NAKAJIMA, A.², KOMORI, Y.³, YOKOKITA, T.³, MORI, D.³, HABA, H.³, YOKOYAMA, A.¹

(¹Kanazawa University, ²University of Tsukuba, ³RIKEN)

^{237}Np ($t_{1/2} = 2.1 \times 10^6$ y), which is produced in nuclear facilities and needs to be investigated for environmental pollution, is expected to be analyzed by accelerator mass spectrometry (AMS). The measurement of ^{237}Np by AMS needs ^{236g}Np ($t_{1/2} = 1.5 \times 10^5$ y) as a spike. Our research group has collected the reaction cross sections in the $^{232}\text{Th} + ^7\text{Li}$ reaction as a fundamental data for the spike production. As well as complete fusion reaction (CF), incomplete fusion reaction (ICF) occurs especially in the reaction of ^7Li which splits into α and t and only a part of the projectile fuses with the target nucleus. In this study, the excitation function of Pa, a product of ICF, was measured, and the mechanism of ICF was discussed by comparing the experimental values with those calculated by code EMPIRE. In addition, we estimated the total reaction cross sections of Pa and Np formation and investigated the effect of incomplete fusion reaction on the whole reaction.

1A03 A recoil correction method of fission cross section measurement in the $^{232}\text{Th} + ^7\text{Li}$ reaction

MORITA, R.¹, TERANISHI, K.¹, HAYAKAWA, Y.¹, SAKAGUCHI, A.², NAKAJIMA, A.²,

KOMORI, Y.³, YOKOKITA, T.³, HABA, H.³, YOKOYAMA, A.¹

(¹Kanazawa Univ., ²Univ. of Tsukuba, ³Riken.)

We are much interested in the $^{232}\text{Th} + ^7\text{Li}$ reaction owing to a possible candidate for the production of ^{236g}Np . The nuclide is expected to be produced mainly at incident energies less than 40 MeV. We aim to measure the fission cross sections at the relevant energies as fundamental data. However, catcher foils for fission fragments make much energy loss of Li projectiles and prevent one from obtaining several data points in one stack at the same irradiation. In this study, we derived a formula for the probability of fragments recoiling out from targets. Comparing the calculated data with the recoil data we measured in the same system, apparent fission cross section for each foil in a stack was corrected. The method was found to be useful not only in obtaining several data in one stack irradiation, but also in reducing the cost of experiment.

1A04 Production of ^{225}Ac in the $^{232}\text{Th}(^{14}\text{N}, xnyp)^{225}\text{Ac}$ reaction

YIN, X., NAMBU, A., KOMORI, Y., MORI, D., HABA, H.

(¹RIKEN Nishina Center)

^{225}Ac is one of the most promising alpha-particle emitting radionuclides for targeted radionuclide therapy. However, a stable supply system of ^{225}Ac has not been established yet in Japan even at the basic research scale (~100 MBq). In this work, we investigated the production of ^{225}Ac in the $^{232}\text{Th}(^{14}\text{N}, xnyp)^{225}\text{Ac}$ reaction by the RIKEN multitracer technique. Production cross sections of ^{225}Ac and its precursor ^{225}Ra were determined at 132 and 116 MeV/u by irradiating metallic ^{232}Th targets ($69 \text{ mg/cm}^2 \times 6$) and ^{27}Al energy degraders ($415 \text{ mg/cm}^2 \times 2$) with a 132 MeV ^{14}N beam (20 pA) extracted from RIKEN Ring Cyclotron. The cross sections were discussed by comparing with those calculated by the PHITS (Particle and Heavy Ion Transport code System) code. Based on our typical experimental conditions (incident beam energy: 132 MeV; beam intensity: 1 μA ; target thickness: 4.5

g/cm²; irradiation time: 2 days), it was estimated that 150 MBq of ²²⁵Ac can be produced at the end of the irradiation. In the symposium, the chemical separation procedure of ²²⁵Ac and the quality of the purified ²²⁵Ac will also be presented.

1A05 Astatine chemistry elucidated in the purification of no-carrier-added astatine by dry distillation

NISHINAKA, I.^{1,2}, HASHIMOTO, K.¹, ISHIOKA, S.N.², WATANABE, S.², SASAKI, I.², AZIM, M.A.³

(¹QST Tokai, ²QST Takasaki, ³BAEC NINMAS) ²¹¹At was produced in the ²⁰⁹Bi(⁴He, 2n)²⁰⁹Bi reaction. The yield was separated from the irradiated bismuth target by a simple method, based on dry distillation. The optimized conditions of the method were studied by monitoring the astatine radioactivity with CdZnTe gamma-ray spectrometers. Dry distillation was accomplished within 6 min of heating the bismuth targets at the bottom of the test tube with an electric furnace, at 650 °C. Cooling the middle of the test tube enabled the recovery of ²¹¹At radioactivity in high yields. The no-carrier-added ²¹¹At solutions were obtained in high yields (26-75%.) by rinsing with the utilization 1.6 mL of eluents, such as distilled water, ethanol, methanol, or chloroform.

Thin-layer chromatography (TLC) analysis revealed that the astatine forms stable anions, At⁻, AtO₃⁻, and AtO₄⁻, in ethanol and aqueous solutions, and these anions are readily oxidized and/or reduced between them in dynamic equilibria owing to the more electropositive character of astatine than other halogens. High-performance liquid chromatography (HPLC) identified large amounts of AtO₃⁻ and At⁻, with lower oxidation states. The observation of different composition profiles by TLC and HPLC suggests that the TLC results were different from the composition profiles of astatine species.

1A06 Production of an atomic beam by ion-electron recombination for atomic-beam spectroscopy of superheavy elements

SUZUKI, H.^{1,2}, ITO, Y.², SATO, T.^{1,2}, AOKI, R.^{2,3}, TSUKADA, K.², ASAI, M.², NAGAME, Y.²

(¹Graduate school of science and engineering, Ibaraki Univ., ²ASRC JAEA, ³Department of science, Ibaraki Univ.)

In the heavy element region, there is a possibility of violation of periodicity on electronic structure of an atom by the influence of the relativistic effect. Especially, focusing on the ground-state electronic configuration of Lr (Z = 103), the valence electron orbital takes 7p_{1/2} rather than 6d_{3/2} assumed from the periodic rule. Our group has succeeded in measuring the first ionization potential of Lr in recent years and obtained a strong support for the 7p_{1/2}[1].

In this work, we have developed an ion-electron recombination atomic beam source for atomic beam spectroscopy. An ion-electron recombination has an advantage of producing a low-energy atomic beam and small chemical dependence. In this system, the ion beam is adjusted by ion optics and injected into the high-density electron cloud. Then, it is recombined with electrons to produce an atomic beam.

In our experiments, the neutralization was indirectly evaluated by measuring the decay of the ion beam current using Rb⁺ beam. As a result, nearly 100 % reduction of the ion beam current through the center of the electron cloud was observed. After improving the ion optics, we are developing the detector for direct measurement of atom. In the presentation, we will report on the experimental results and the future prospects.

[1] T.K. Sato, et al., *Nature* **520** 209 (2015).

1A07 Direct mass measurement of superheavy nuclei ²⁵⁷Db via MRTOF+α-TOF

NIWASE, T.^{1,2,3}, SCHURY, P.³, WADA, M.³, BRIONNET, P.², CHEN, S.⁴, HASHIMOTO, T.⁵, HABA, H.², HIRAYAMA, Y.³, HOU, D.S.^{6,7,8}, IIMURA, S.^{9,2,3}, ISHIYAMA, H.², ISHIZAWA,

S.^{10,2}, ITO, Y.¹¹, KAJI, D.², KIMURA, S.², KOURA, H.¹¹, LIU, J.J.⁴, MIYATAKE, H.³, MOON, J.Y.⁵, MORIMOTO, K.², MORITA, K.^{1,2}, NAGAE, D.¹, ROSENBUSCH, M.², TAKAMINE, A.², WATANABE, Y.X.³, XIAN, S.X.⁴, YAN, S.X.¹², WOLLNIK, H.¹³

(¹Kyushu Univ., ²RIKEN, Nishina Center., ³KEK., ⁴HongKong Univ., ⁵IBS., ⁶IMP., ⁷University of Chinese Academy of Science, ⁸Lanzhou Univ., ⁹Osaka Univ., ¹⁰Yamagata Univ., ¹¹JAEA, ¹²Jinan Univ. ¹³NMSU)

The atomic mass is an important quantity that faithfully reveals the interactions between nucleons of each nuclide. According to the liquid drop model, superheavy elements cannot exist due to the fission barrier. Therefore, their existence is permitted solely by the shell effect, and mass is an exclusive quantity that can explore for shell effect of each nucleus.

Previously, the mass of heavy nuclides including transfermium isotopes produced in fusion reactions has been measured by using an MRTOF-MS combined with GARIS-II. Recently, a novel α -TOF detector has been developed. It is possible to measure mass and correlated decay properties. The decay signal as a fingerprint of superheavy nuclei also enables highly accurate measurement to distinguish from backgrounds with just a few events per day.

Experiments were performed with RIKEN Ring Cyclotron. We succeeded the first direct mass measurement of the superheavy element ²⁵⁷Db isotopes, produced in the ²⁰⁸Pb(⁵¹V, 2n) reaction, by use of the MRTOF-MS equipped with an α -TOF. We observed 11 events with times-of-flight consistent with that expected for ²⁵⁷Db followed by the detection of an alpha-decay consistent with ²⁵⁷Db or one of its decay products. In this presentation, we will report the details of the experiment and result.

1A08 Development of an RF ion-collection system for nuclear-chemistry studies

SHIGEKAWA, Y.¹, YAMAGUCHI, A.¹, SATO, N.¹, TAKAMINE, A.¹, WADA, M.², HABA, H.¹
(¹RIKEN, ²KEK)

The RF ion-collection apparatus, consisting of a gas cell and an RF carpet, allows the rapid deceleration and transportation of high-energy RI ions, and has been used for nuclear-physics studies such as precision mass measurement of heavy nuclei. Applying this type of apparatus to nuclear-chemistry studies enables new studies such as gas-phase chemistry of short-lived superheavy elements (half-life <1 s). In this study, we developed a new RF ion-collection apparatus for nuclear-chemistry studies, and evaluated ion extraction time and efficiency. The constructed apparatus includes a cryogenic He gas cell, where purified He gas is introduced; the cooling and purification systems reduce impurities and prevent ions from reacting with impurities. The ions decelerated inside the gas cell are transported to an RF carpet with a DC electric field gradient, then to the central hole of the carpet with the RF-surfing technique, and subsequently to a high vacuum through a quadrupole ion guide. To evaluate ion extraction time and efficiency, we extracted ²²⁰Rn ions recoiling out of a ²²⁴Ra source at various conditions. We determined the maximum extraction efficiency to be 60–70%. The extraction times were ~20 and ~30 ms at 100 and 200 mbar of He gas, respectively (the pressures are obtained by converting the pressures at low temperature to the ones at room temperature). The developed apparatus allows efficient extraction of ions with half-lives >100 ms, leading to nuclear-chemistry studies using short-lived RIs that have been difficult to study thus far.

1A09 Half-life determination of nuclear excited states of ²²⁹Th by the α - γ coincidence measurement of ²³³U

SHIGEKAWA, Y.¹, YAMAGUCHI, A.¹, SUZUKI, K.², HABA, H.¹, HIRAKI, T.², KIKUNAGA, H.³, NISHIMURA, S.¹, SASAO, N.², YOSHIMI, A.², YOSHIMURA, K.²
(¹RIKEN, ²Okayama Univ., ³Okayama Univ., ³Tohoku Univ.)

The first nuclear excited state of ^{229}Th , $^{229\text{m}}\text{Th}$, has an excitation energy as low as 8.3 eV, which may allow nuclear half-life variation depending on chemical environments and realization of an ultraprecise nuclear clock. Although the internal conversion process of $^{229\text{m}}\text{Th}$ has been recently observed, the γ -rays from $^{229\text{m}}\text{Th}$ have never been observed clearly. Elucidating the properties of highly excited states of ^{229}Th is important to estimate the half-life of the γ -ray emission of $^{229\text{m}}\text{Th}$, which helps us to measure the γ -rays from $^{229\text{m}}\text{Th}$. In this study, we measured the half-lives of some highly excited states of ^{229}Th through α - γ coincidence measurement of ^{233}U . A ^{233}U source prepared by electrodeposition was placed in a vacuum chamber. The detection timing and energy of α particles from the source were measured using a Si detector, while those of γ rays were measured using a $\text{LaBr}_3(\text{Ce})$ scintillator. From the 4784-keV α particles and the 42.4-keV γ rays, we determined the half-life of the 42.4-keV state to be 171(4) ps, which is consistent with the literature value. The half-life determined for the 97.1-keV state was different from the literature value. We successfully determined the half-lives of the 163.2-keV and 71.8-keV states for the first time. Based on the obtained half-lives, we will estimate and report the half-life of the γ -ray emission of $^{229\text{m}}\text{Th}$.

1A10 Discovery of $^{234\text{m}}\text{Np}$ isomer and its decay properties

ASAI, M.^{1,2}, SUEKAWA, Y.², HIGASHI, M.², KAMADA, H.³, TOBE, T.⁴, ANDREYEV, A.N.^{1,5}, HIROSE, K.¹, ITO, Y.¹, MAKII, H.¹, NISHIO, K.¹, NIWASE, A.², ORLANDI, R.¹, SAKAMA, M.⁶, SATO, T.K.¹, SHIBATA, M.⁷, SUZAKI, F.¹, SUZUKI, H.¹, TOKOI, K.¹, TOMITSUKA, T.¹, TSUKADA, K.¹
(¹JAEA, ²Kyushu Univ., ³Nagoya Univ., ⁴Kyoto Sangyo Univ., ⁵Univ. York, ⁶Tokushima Univ., ⁷Nagoya Univ. RI)

A new isomer $^{234\text{m}}\text{Np}$ with a half-life of ~9 min has been discovered. It was produced in the reaction of ^{233}U target with ^6Li beam at JAEA tandem accelerator, and was identified through

characteristic X-ray measurement and mass separation with an on-line isotope separator (ISOL). Gamma-ray singles, gamma-gamma coincidence, and internal-conversion electron measurements were performed. It was revealed that the $^{234\text{m}}\text{Np}$ decays mostly through an isomeric E4 transition followed by an M1 transition, and that the spin-parity of the $^{234\text{m}}\text{Np}$ should be 5^+ . The results are introduced in detail, and the nuclear structure of the $^{234\text{m}}\text{Np}$ are discussed.

1B01 A thermodynamic study on the complexation of actinides with transferrin

SUZUKI, T., KOBAYASHI, T., SASAKI, T.
(Kyoto Univ.)

In the present work, we focused on the complexation of actinides with transferrin. Transferrin known as a transporter of ferric ions in human blood can play an important role in in-vivo kinetics of actinides. However, a detailed complexation mechanism of actinides with the coordination sites of transferrin is not well known. Thermodynamic data of the complexation of actinides and transferrin would be important when considering an elimination of actinides from blood by using chelating medicines. In this work, complexations between transferrin and uranium (U(VI)), thorium (Th(IV)) and zirconium (Zr(IV)) as a chemical analog of plutonium were investigated by UV-Vis spectrometry.

Aliquots of U(VI) or Zr(IV) carbonate stock solution equivalent to 0.2~10 mole were added into transferrin solution in constant concentration (0.01~0.06 mM). Aliquots of thorium NTA stock solution were also prepared and mixed with the transferrin stock solutions.

An absorption peak at 240~250 nm appeared in the mixture of U(VI) and transferrin. Under the constant metal-transferrin ratio, an absorption peak at 240~250 nm appeared and its intensity depended on the total transferrin concentration with a gradual shift of the peak position from 240 to 250 nm. Therefore, it seems that coordination

structures of metal complexes are different by total transferrin and metal concentration.

1B02 Development of a rapid analytical method for U, Np, and Pu using ICP-MS and ICP-MS/MS in urine samples for patient screening after radiation accident

YANG, G., ZHENG, J., KIM, E., SENO, H., KOWATARI, M., KURIHARA, O.

(National Institutes for Quantum and Radiological Science and Technology)

Internal contamination with alpha-particle emitting actinides, such as ^{238}U , ^{237}Np , ^{239}Pu , ^{240}Pu , is likely to bring a large radiation exposure dose to the patient even if the intake amount is small. In order to provide timely information for prompting decision making in radiation emergency medicine, we attempted to develop a simple and rapid method based on the analysis of ultra-trace level of U, ^{237}Np and Pu isotopes in a small amount of urine samples (20–100 mL) by the SF-ICP-MS and the ICP-MS/MS. A simple single chromatographic column packed with 2 mL AG MP-1M resin (100–200 mesh, chloride form, Bio-Rad, Hercules, CA, USA) was used for the separation of targeted isotopes from the matrix elements and other potential interferences. Uranium isotopes were measured using ^{233}U as a yield tracer with a yield of ca. 10%. To avoid polyatomic and tailing effects from U, ^{237}Np and Pu isotopes were collected after U fraction and measured using ^{242}Pu as a yield tracer with a yield of ca. 70%. Finally, 4 urine reference materials with Pu addition and 3 urine reference materials with U addition provided by the Association for the PROMotion of Quality COntrol in RADiotoxicological Analysis, France were analyzed by the proposed method and the conventional alpha spectrometry for validation.

1B03 Molecular-weight-dependent distribution of humic acid induced by gamma-irradiation complexed with metal ions

ZHAO, Q., KOBAYASHI, T., SASAKI, T.
(Kyoto Univ.)

Gamma-irradiation field generated by radioactive wastes has been recognized to affect the interaction of the radionuclides leached out from the radioactive waste with humic acid in natural groundwater. The molecular weight of the complexes would be critical for the environmental migration of radionuclides. In the present study, the effect of gamma-irradiation on the chemical properties of humic acid (HA) fractions, and the molecular-weight distribution of metal ions were investigated. The gamma-irradiated humic acid fraction was obtained by ultrafiltration into four fractions (> 100, 50-100, 10-50, < 10 kDa). The experimental results from total organic carbon measurement and potentiometric titration suggested that some of irradiated HA molecules were degraded to the smaller molecular weight, and phenolic –OH would be a predominant functional group of the small-molecular-weight fractions after the radiolysis degradation. When an aliquot of Cs, Sr, Eu (as an analog of trivalent actinides) stock solution was added to the HA solutions, it is found that more metal cations are gradually belonging to the smaller molecular-weight fractions with dose.

1B04 Structure of ruthenium complex in nitric acid solution estimated using DFT calculation

KATO, A.¹, KANEKO, M.², NAKASHIMA, S.^{1,3,4}

(¹Grad. School of Sci., Hiroshima Univ., ²Japan Atomic Energy Agency, ³N-BARD, Hiroshima Univ., ⁴Grad. School of Advanced Sci. and Eng., Hiroshima Univ.)

The Ru atom exists as trivalent nitrosyl-ruthenium ion, $[\text{Ru}(\text{NO})]^{3+}$, in nitric acid solution such as high-level liquid waste and forms the complex with H_2O and NO_3^- ligands. The composition of the coordination species varies depending on the concentration of nitric acid. In the previous experiment reports of $[\text{Ru}(\text{NO})(\text{NO}_3)_x(\text{H}_2\text{O})_y]^{(3-x)+/-}$ ($x + y = 5$ for unidentate NO_3^- coordination) system, the complexes with $x = 0-4$, especially,

$[\text{Ru}(\text{NO})(\text{NO}_3)_3(\text{H}_2\text{O})_2]$ were observed depending on the concentration of nitric acid. However, such systematic properties of the nitrosyl-ruthenium complexes have not been understood, although the precise modeling of coordination geometries in nitric acid solution is required to understand the separation mechanism in solvent extraction. Therefore, we performed density functional theory calculation to the nitrosyl-ruthenium complexes, which included the 12 patterns of geometrical structures of $[\text{Ru}(\text{NO})(\text{NO}_3)_x(\text{H}_2\text{O})_{5-x}]^{(3-x)+/-}$ ($x = 0-5$), for modeling the stable coordination structures and complexation reactions of NO_3^- ligands toward $[\text{Ru}(\text{NO})(\text{H}_2\text{O})_5]^{3+}$. The complexation reaction with nitric acid proceeding to $[\text{Ru}(\text{NO})(\text{NO}_3)_3(\text{H}_2\text{O})_2]$ was confirmed by introducing the effect of outer-sphere molecules. In addition, the geometrical isomer, in which NO_3^- was coordinated only to the equatorial position of NO, was expected to be the most stable structure by using electron density analyses.

1B05 Property of Dimetallofullerene Encapsulating Promethium

SUWA, T.¹, AKIYAMA, K.¹, KIKUNAGA, H.², KUBUKI, S.¹

(¹Grad. School Sci., Tokyo Metropolitan Univ., ²ELPH, Tohoku Univ.)

In recent years, it has been reported that the stability of dimetallofullerenes (di-EMF) with two lanthanide atoms decreases with the increase in the atomic number of encapsulated lanthanides, and also reported that the di-EMFs with heavy lanthanides are stabilized by the reduction of di-EMFs. In our previous work, we could not have observed any promethium di-EMF species under the atmospheric condition. In this work, we reviewed the present scheme of metallofullerene separation/extraction and tried to develop the extraction apparatus under anaerobic condition. About 1 g of Sm_2O_3 pellet was irradiated with 50 MeV bremsstrahlung radiation for 6 hours. Produced promethium (Pm) was separated from this irradiated Sm pellet by the chemical

separation. The Pm tracer with a nitrate form was dissolved to ethanol and mixed with Ce-139 radiotracer and La nitrate as a carrier. These Pm solutions were adsorbed on a porous carbon rod and sintered. These Pm carbon rods were set into a fullerene generator and then discharged under He atmosphere. Produced soot containing Pm di-EMFs were refluxed under anaerobic condition by using newly developed fullerene extraction apparatus in this work. As a result, the radioactivity of extracted Pm under anaerobic condition was found to be 3.3 times larger than that under atmospheric condition.

1B06 Development of mass spectrometric method for simultaneous determination of hard-to-measure Pu isotopes in environmental and reactor decommissioning waste samples

ZHENG, J.¹, WANG, H.², SHIKAMORI, Y.³, NI, Y.⁴, AONO, T.¹

(¹National Institutes for Quantum and Radiological Science and Technology, ²Univ. South China, ³Agilent Technol., ⁴Peking Univ. China)

Mass spectrometry, especially ICP-MS, as an atom-counting approach, is gradually replacing alpha spectrometry as a main-stream technique for Pu isotopic analysis. However, as Pu is often concomitant with a large (5-10 orders of magnitude) excess of U in environmental matrix, which clearly obscures the determination of ²³⁸Pu using any mass spectrometer due to the isobaric interference from ²³⁸U. So far, there is no established mass spectrometric method for simultaneous determination of these hard-to-measure ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu and ²⁴¹Pu in environmental mediums. In this study, we developed an advanced mass spectrometry method for simultaneous analysis of important Pu isotopes (^{238,239,240,241}Pu) using triple quadrupole ICP-MS/MS by employing the ion-molecule reaction separate ²³⁸U and ²³⁸Pu, which was previously difficult to be measured. By introducing a desolvation sample introduction system, we succeeded in increasing 10 times the sensitivity of Pu isotopes and suppressing the

production of uranium hydride, we achieved the detection limit of fg level in on-mass mode. We have developed a new low-temperature fusion method for environmental and nuclear decommissioning samples. By combining these new sample preparation techniques with the developed ICP-MS/MS detection, rapid Pu isotope mass spectrometry methods for environmental samples and decommissioning related waste samples were established.

1B07 Trace element composition of radioactive particles

SUEKI, K.¹, HASEGAWA, R.², MATSUO, K.², ISHII, T.², ADACHI, K.³, SATOU, Y.⁴

(¹Faculty Pure Applied Sci., Univ. Tsukuba, ²Grad. Sch. Pure Applied Sci., Univ. Tsukuba, ³MRI, ⁴JAEA CLADS)

There are radioactive particles that are insoluble in water and are mainly composed of silicate, released by the Fukushima Daiichi Nuclear Power Plant accident. Radioactive particles are known to be roughly divided into two types and are characterized by the ¹³⁴Cs/¹³⁷Cs radioactivity ratio. In this study, we carried out total decomposition of type B radioactive particles by mixed acid, and measured multi-element quantitative analysis by triple quadrupole inductively coupled plasma mass spectrometry (ICP-QQQ-MS). It isn't possible to analyze the silicon and the oxygen which are a main element of a radioactive particle by ICP - QQQ - MS. We added a result of the elemental analysis of a radioactive particle from a result of SEM/EDS about several elements and got the whole analytical value. When correlation with Si was checked, many elements had positive correlation, but the coefficient of correlations of the several elements (As, Se, Ag, Cd, Sn, Sb, Cs, Ba, Pb, and U) were not good. The ¹³³Cs/¹³⁷Cs ratio ranged from 1 to 5, especially 6 out of 10 radioactive particles were about 1, which was close to the ratio that would be encountered in ²³⁵U neutron induced fission. The included amount of U showed with 1.0×10^{-3} - 4.6 ng. In particular, the particle which included 4.6 ng indicated 10 to

100 times compared with other particles. We expect to contribute to guess at the process of the radioactive particle production as a result of the minor element constitution of obtained particles.

1B08 Fluorescence property of radiocesium-bearing microparticles emitted from Fukushima Daiichi Nuclear Accident

ABE, Y.^{1,2}, ONOZAKI, S.², ISHIKAWA, M.¹, NAKAI, I.¹, KITA, K.³, IGARASHI, Y.^{3,4}, OURA, Y.⁵, TSURUTA, H.⁶, MORIGUCHI, Y.⁷
(¹Tokyo Denki Univ., ²Tokyo Univ. Sci., ³Ibaraki Univ., ⁴KURNS, ⁵Tokyo Met. Univ., ⁶RESTEC, ⁷Univ. Tokyo)

Considerable amounts of radioactive materials were released into the environment following the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011. As a form of radioactive materials emitted from the FDNPP, radiocesium-bearing microparticles (CsMPs) have been investigated by numerous researchers. In this study, we focused on Type A of CsMP which was widespread over the Eastern Japan at the early stage of the accident, and investigated its fluorescence property with the aim to propose the rapid and easy detection method of CsMPs from environment. We isolated some Type A CsMPs from aerosol samples collected just after the accident and measured fluorescence spectrum and fluorescence distribution of individual particles using a multi-photon excitation microscope. As the result, this study first demonstrated that CsMPs emit characteristic green fluorescence in response to certain wavelengths of light. X-ray analyses of CsMPs using synchrotron radiation X-ray microbeam were carried out at SPring-8 and revealed that they contain a trace amount of U in the form of UO₂²⁺ ion. We thus concluded that the fluorescence from Type A CsMP originate to UO₂²⁺ ion, one of well-known strong fluorescence species in silicate glass matrix.

1B09 Application of Kelvin Probe Force Microscopy to the measurement of surface

potential of radiocaesium-bearing microparticles

OKUDA, T.¹, SHINKE, Y.¹, KUROSAWA, K.¹, IWATA, A.¹, SATOU, Y.², ABE, Y.³, IGARASHI, Y.⁴

(¹Keio Univ., ²JAEA, ³Tokyo Denki Univ., ⁴Kyoto Univ.)

The Fukushima Daiichi Nuclear Power Plant accident caused serious emission of radionuclides into the atmosphere. After several years, insoluble radiocaesium-bearing microparticles (CsMP) have been found in some regions in Japan. These insoluble solid particles remain as a particle in human airways and continue emitting radiation when they are inhaled. Therefore, these particles have a potentially significant adverse effect on human health. Because of the continuous emission of the β -rays, which is energetic electron beam, previous studies reported that the radioactive particles would have specific electrostatic charging characteristics. Also, several laboratory experiments and numerical simulations have shown that the electrostatically charged particles are more deposited on the human respiratory airway. Therefore, the electrostatic charging characterization of the CsMP is important for the accurate evaluation of its health effects. In this study, we used Kelvin Probe Force Microscopy to measure the surface potential of CsMP. Charge number controlled NaCl particles were also measured to calibrate the surface potential determined by KPFM and the actual charge number. As a result, linear relationship between charge number and surface potential were obtained.

1B10 Analysis and simulated generation of the radioactive particles emitted from the Fukushima nuclear accident

INAGAKI, M.¹, SATO, R.², SEKIMOTO, S.¹, TAKAMIYA, K.¹, OKI, Y.¹, OHTSUKI, T.¹

(¹KURNS, ²Kyoto Univ.)

After the Fukushima Daiichi Nuclear Power Plant accident, various kinds of radioactive materials were released to the surrounding

environment. Among those materials, large amounts of glassy particles have been found. To study physical and chemical properties of those glassy particles can make us understand what happened in the nuclear reactors during the accident. Therefore, we generated the simulated glassy particles, and aimed at elucidation of the formation process of the particles. In this study, we analyzed the actual glassy particles and conducted the simulated generation of the particles. As a result of SEM observation of the actual glassy particles, diatomaceous earth was discovered from the surfaces of the particles. Diatomaceous earth is a strong candidate for the source material of the particles. The simulated particles were produced by heating a mixture of diatomaceous earth and artificial seawater. Then, the simulated particles generated were studied by SEM-EDS analysis. As a result of the analysis, the simulated particles were similar to the actual glassy particles. One hypothesis is that diatomaceous earth and seawater that existed in the reactor at the accident were heated, and radioactive materials were taken in and the glassy particles were generated.

2S01 Studies on Transfer Parameters in the Living-sphere for Estimation of Radiation Dose to Humans from Environmental Radionuclides

TAGAMI, K.

(Natl. Inst. Quantum Radiol. Sci. Technol.)

One important purpose to study transfer of radionuclides in the environment is to assess the radiation effects to human beings. Accurate measurements of low-level radionuclides in the environment are therefore necessary; “radiochemistry” is a strong tool for these works. Besides such data in environmental materials can provide useful information, that is, transfer parameters to express the ratio of radionuclide transfer rate from one compartment to the other, i.e., from soil to plant. Parameter values have been used in mathematical models for dose assessments; because we know that parameter values have ranges so single datum is not trustful.

Thus collating parameter data as many as possible by measuring of environmental samples, doing tracer experiments and surveying radiochemically sense literature values needs to be carried out to make the dose estimation results more accurate. I have been involved to measure and collate parameter values obtained in Japan and that experience was highly helpful when the Fukushima Daiichi Nuclear Power Plant accident occurred. After the accident, using many measurement results of environmental samples, we could provide transfer parameter values. At the presentation, I will summarize our almost 10 years study results related to the accident.

2A01 Coprecipitation experiment of alkaline earth elements with Barium sulfate, towards the experiment of element 102, No.

HAYAMI, S.¹, KASAMATSU, Y.¹, WATANABE, E.¹, NAKANISHI, R.¹, TONAI, K.¹, TAKAMIYA, K.², SHINOHARA, A.¹

(¹Grad. Sch. of Sci., Osaka Univ., ²KURNS, Kyoto Univ.)

Nobelium (¹⁰²No) belongs to an actinide series. It is reported that stable oxidation state of No is +2 although those of other heavy actinide elements are all +3. The chemical behavior of No in HCl was reported to be similar to that of Group 2 elements. Our group has investigated coprecipitation of No with the Sm hydroxide. For newly investigating the chemical behavior of the divalent elements and nobelium, we focused on the reaction with sulfate ion.

In this study, we investigated sulfate coprecipitation behavior for alkaline earth elements using ⁴⁷Ca, ⁸⁵Sr, ¹³³Ba, and ²²⁶Ra tracers and discussed the applicability of this method to nobelium experiment. As a result, clear differences in coprecipitation behaviors among these elements were observed. The tendency of the sulfate coprecipitation yields for the elements is consistent with the order of their solubility products. We also found that there are no clear differences between coprecipitation and precipitation behaviors of alkaline earth elements. Based on these results, we concluded that this

method is able to be applied to the experiment of No.

2A02 On-line Isothermal Gas Chromatographic Behavior of Oxychlorides of Group-5 Elements, Nb, Ta, and Db

SATO, T.K.^{1,2}, CHIERA, N.M.^{1,3}, TOMITSUKA, T.¹, TOKOI, T.⁴, SUZUKI, H.^{1,2}, ITO, Y.¹, ASAI, M.¹, TSUKADA, K.¹, SHIRAI, K.⁵, INOUE, H.⁵, ADACHI, S.⁶, KASHIHARA, A.⁶, MAKII, H.¹, HIROSE, K.¹, NISHIO, K.¹, WATANABE, E.⁴, SAKAMA, M.⁷, GOTO, S.⁵, NAGAME, Y.¹

(¹Japan Atomic Energy Agency, ²Ibaraki Univ., ³Paul Scherrer Institute, ⁴Osaka Univ., ⁵Niigata Univ., ⁶Univ. of Tsukuba, ⁷Tokushima Univ.)

The influence of strong relativistic effects on chemical properties has been interesting in the superheavy element region. Their chemical properties, however, have not been investigated sufficiently because of experimental difficulties owing to their low production rates and short half-lives. In order to elucidate the chemical properties of dubnium (Db, Z = 105), we have conducted on-line isothermal gas chromatographic experiments of oxychloride of group-5 elements. We confirmed the formation of volatile oxychlorides of Db and its lighter homologs Nb and Ta by using ²⁶²Db (half-life, $T_{1/2} = 33.8$ s), ⁸⁸Nb ($T_{1/2} = 14.5$ min.), and ¹⁷⁰Ta ($T_{1/2} = 6.76$ min.), respectively. We successfully determined the adsorption enthalpies of the oxychlorides of each element on the quartz surface from their isothermal gas chromatographic behavior. The obtained volatility sequence of the group-5 elements is found to be Nb > Ta ≥ Db.

2A03 Anion-exchange behavior of Db in HF/HNO₃ solution

KATO, M.¹, ADACHI, S.¹, TOYOSHIMA, A.², TSUKADA, K.³, ASAI, M.³, HABA, H.⁴, YOKOKITA, T.⁴, KOMORI, Y.⁴, WANG, Y.⁴, SHIGEKAWA, Y.⁴, MORI, D.⁴, KASHIHARA, A.¹, NAKAJIMA, A.¹, SUZUKI, Y.¹, NISHIZUKA, K.¹, TOKOI, K.², SUEKI, K.¹

(¹Univ. of Tsukuba., ² Osaka Univ., ³JAEA, ⁴RIKEN)

A few studies have been so far performed on aqueous chemistry of Db, the group-5 superheavy element, by anion-exchange experiments in HF and HF/HNO₃ mixture solutions. Although the distribution coefficient (K_d) of Db showed the trend of Ta>Nb>Db>Pa in the group-5 homologs and pseudo-homolog elements, its chemical species has not been clarified. In this work, for identification of fluoride species of Db, we performed anion-exchange experiments of Nb, Ta, and Db in HF/1.0 M HNO₃ mixture solutions by using an automated rapid chemistry apparatus, ARCA.

^{88g}Nb, ¹⁷⁰Ta, and ²⁶²Db were produced in the ^{nat}Ge(¹⁹F, xn)^{88g}Nb, ^{nat}Gd(¹⁹F, xn)¹⁷⁰Ta, and ²⁴⁸Cm(¹⁹F, 5n)²⁶²Db reactions, respectively, at the RIKEN K70 AVF cyclotron. The reaction products, transported by a He/KCl gas-jet system, were dissolved with HF/1.0 M HNO₃ mixture solution at a flow rate of 1.0 mL min⁻¹. For Db, after elution of HF/1.0 M HNO₃ from the anion-exchange column filled with CA08Y resin, the 2nd stripping solution was fed into the column. We measured α -particles for dried samples, which were prepared from the above-mentioned eluents, by using an automated rapid α /SF detection system.

We evaluated the K_d values of Nb and Ta obtained in the online column experiment. The results were almost the same as those of the batch experiment performed previously. In the Db experiments, we repeated anion exchange experiments 350 times and observed alpha events of ²⁶²Db in the 1st and 2nd fractions, respectively.

2A04 Anion exchange of Rf in H₂SO₄: H₂SO₄ concentration dependence of distribution coefficients

YOKOKITA, T.¹, KASAMATSU, Y.², WATANABE, E.², KOMORI, Y.¹, SHIGEKAWA, Y.¹, MORI, D.¹, WANG, Y.¹, NINOMIYA, H.², HAYAMI, S.², TONAI, K.², GHOSH, K.¹, SHINOHARA, A.², HABA, H.¹

(¹RIKEN, ²Osaka Univ.)

It is difficult to obtain the distribution data at equilibrium for the short-lived ²⁶¹Rf in 2-phase distribution experiments, but such data are necessary to discuss in detail the chemical properties of Rf. To obtain distribution coefficients (K_d) of Rf, a batch-type solid-liquid extraction apparatus called AMBER was developed. We have been studying the sulfate complexation of Rf by an anion-exchange method. In our previous study, we successfully obtained the K_d value of Rf at 0.11 M H₂SO₄. The obtained K_d value of Rf is low ($K_d < 15 \text{ mL g}^{-1}$), probability indicating that Rf does not form anionic species, or counter ions of HSO₄⁻ and SO₄²⁻ are preferentially adsorbed on the anion-exchange resin. In this study, we performed the anion-exchange experiment of Rf at several H₂SO₄ concentrations including the lower concentration of 0.060 M H₂SO₄.

We performed anion-exchange experiments of ²⁶¹Rf and ¹⁶⁹Hf by using AMBER. ²⁶¹Rf and ¹⁶⁹Hf were produced in the ²⁴⁸Cm(¹⁸O,xn)²⁶¹Rf and ^{nat}Gd(¹⁸O,xn)¹⁶⁹Hf reactions, respectively, by using the AVF cyclotron at RIKEN. The products were transported from the target chamber to AMBER with a He/KCl gas-jet system. After the anion exchange with AMBER, radioactivities of ²⁶¹Rf and ¹⁶⁹Hf were investigated by using an automated rapid α /SF detection system and Ge detectors, respectively.

We obtained the K_d value of Rf in 0.060–0.46 M H₂SO₄. The K_d values of Rf are low in all the H₂SO₄ concentrations studied, suggesting that Rf does not form anionic species and the sulfate complexation strength of Rf is lower than those of Zr and Hf.

2A05 The method to reduce “particle mixing” under automated particle measurement (APM) condition of LG-SIMS

TOMITA, R., ESAKA, F., YOMOGIDA, T., MIYAMOTO, Y.
(JAEA)

Large Geometry Secondary Ion Mass Spectrometry (LG-SIMS) is one of the strongest tools for analyzing isotope ratios of micron sized

uranium particle. LG-SIMS has high spatial resolution of less than 1 μm with microprobe mode. However, this capability is bit less under automated particle measurement (APM) condition. If two or more particles are located in a quite narrow area, APM may detect the cluster as one particle. This “particle mixing” effect shows analytical results including false isotope ratios. In order to investigate how often particle mixing happens and how to solve this problem, we implemented APM to mixed uranium particle standard (U010, U100, U350 and U850) and try to apply particle manipulation with APM. In our experiment, each area ($350 \times 350 \mu\text{m}^2$) was scanned with an O^{2+} primary beam with a current of 1.5 nA for 9 sec. Then, secondary ion images were recorded for circular area with a radius of 8500 μm on the center of a sample planchet. The APM detected 5976 particles, and 1943 particles (32%) in them showed false isotope ratios. In addition to “particle mixing”, U850 cluster was shifted down to around 75% enrichment. The sample showed too high hydride rate, so that ^{235}UH interfered ^{236}U . This interference caused false ^{236}U abundance and lead to false ^{235}U abundance. To solve these problems, 50-80 particles were manipulated randomly from another mixed standards planchet and analyzed them by APM.

2A06 An examination of environmental sample analyses for safeguard using multi-collector ICP-MS

TOMITA, J., TOMITA, R., SUZUKI, D., YASUDA, K., MIYAMOTO, Y.

(Japan Atomic Energy Agency)

Multi-collector type ICP-MS (MC-ICP-MS) is a powerful tool to precisely and accurately analyze the amount and isotopic ratios of ultra-trace (fg to ng level) U and Pu in an IAEA environmental safeguards sample. Clean Laboratory for Environmental Analysis and Research (CLEAR) of JAEA installed a MC-ICP-MS instrument, and is preparing the measurement protocol for uranium and plutonium isotope ratios. In this presentation, the accurate measuring method of

uranium isotope ratios with high precision using MC-ICP-MS was examined.

Quantitative contribution of tailing and hydration of major isotopes (^{235}U , ^{238}U) to minor isotopes (^{234}U , ^{236}U) were evaluated by measuring uranium certified reference materials (BNL CRM U015, U0002). The results measured the U0002 solution showed that the ratio of ^{238}U tailing at ^{236}U to $^{238}\text{U}^+$ was 10^{-8} . Relative standard deviations of uranium isotope ratios of U015 were measured to optimize the number of cycles for measuring a sample. The significant differences among the mean values of 5, 10 and 20 cycles were not found for $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$ isotopic ratios. The frequency for the correction of mass bias and ion counter efficiencies for accurate measurements of U isotopic ratios will be discussed in the presentation.

2A07 Composition of Slags Produced from Domestic Waste Determined by Photon Activation Analysis 2

AKIYAMA, K.¹, SAITO, R.¹, SUWA, T.¹, ALI, A.¹, KIKUNAGA, H.², KUBUKI, S.¹

(¹Grad. School Sci., Tokyo Metropolitan Univ., ²ELPH, Tohoku Univ.)

Mineral resource including rare metals are considered to be an extremely important resource that support many industrial fields in the world. It is well known that various mineral resources are imported because of the poor resource in our country. On the other hands, it is also well known that the waste from consumer electronics and mobile phone that we emitted contain a great deal of mineral resources and is called “Urban Mine”. Recently, many researchers and engineers have tried to recycle mineral resources from such urban mines for reducing the amount of the imported rare metals. As a part of such a resource recycling research, we have studied the composition of a domestic waste slag, which is kind of an artificial sand produced from incineration ash of domestic waste in Tokyo, in order to assess the potential for the resource recycling from it.

In this study, the composition of the slag in Sagami-hara City, Kanagawa Prefecture, which produces domestic waste slag as in Tokyo, was studied by using the photon activation analysis, and its results were compared with those in Tokyo.

2A08 Variability in Composition of Domestic Waste Slag and Separation of Valuable Metal Elements from them

SAITOU, R.¹, AKIYAMA, K.¹, ALI, A.S.A.¹, KHAN, I.¹, KIKUNAGA, H.², KUBUKI, S.¹
(¹Grad. School Sci., Tokyo Metropolitan Univ.,
²ELPH, Tohoku Univ.)

Domestic waste slag is a kind of artificial sand produced by the melt/quenching method of the incineration ash from the combustible waste. It has been considered that these slag contain several kinds of valuable metal elements, such as Cu and Au, and are important materials from the view point of resource recycling. In our previous work, we reported the composition of monthly provided slag from January 2018 to December 2018 by photon activation analysis (PAA). In this work, we will report the composition of monthly provided slag in 2019 by PAA and XRF and also, we discuss about the possibility of component separation by magnets.

Monthly collected domestic waste slag (Slag 1901-1912) at the Tamagawa Incineration Plant in 2019 was pulverized and wrapped by high pure Al foil. These slag samples were irradiated with 20 MeV bremsstrahlung for 6 hours in ELPH at Tohoku University. After the irradiation, γ ray emitted from slag samples were determined by a high purity Ge semiconductor detector.

As results, the averaged main composition of slag in 2019 was found to be almost same as that in 2018.

2A09 Activation analysis of shells of Japanese basket clams (shijimi)

REZA, Md.S., OURA, Y.
(Tokyo Metropolitan Univ.)

Shells of Japanese basket clams (shijimi) living in brackish lakes were analyzed by instrumental

photon activation analysis (IPAA) and instrumental neutron activation analysis (INAA). A single comparator method was applied in IPAA. About 10 elements were detected by INAA and IPAA each. As, C, Rb, Y, and Zr were detected by only IPAA. Elemental contents in 2 shells of an individual were almost consistent. Among different individuals most of elemental contents also consistent, but especially, some individuals have higher content for Mn. Shells from four different locations were also analyzed. Major and minor elements were contained in similar concentration even for different locations. But for some trace elements, especially Mn, the concentrations varied among production areas. No correlations between elemental concentrations and locations were observed.

2A10 Determination of elements in sediments of lakes in Gunma prefecture by neutron activation analysis

OKADA, Y.¹, KUMAGAI, N.¹, WATANABE, S.², NOHARA, S.³
(¹Tokyo City Univ., ²Gunma Prefectural Fisheries Experiment Station, ³National Institute for Environmental Studies)

In Akagi-Onuma, Gunma Prefecture, the concentration of radioactive cesium in the lake water continues to decline gradually. In order to investigate the sediment characteristics of Akagi Onuma, the elements in the sediments of 11 lakes in Gunma prefecture were analyzed by neutron activation analysis. The sediment (0 mm to 40 mm depth) was irradiated with KUR for 30 seconds and 1 hour to quantify the elements. The quantification was performed by the comparison method using rock standard substance JLK1-1. As a result, 25 elements were quantified. All lake sediments contained more than 20% Si and a few% Al and Fe. Stable Cs content is 1.4 $\mu\text{g} / \text{g}$ in Lake Haruna, 2.8 $\mu\text{g} / \text{g}$ in Lake Narusawa and Usui, 2.9 $\mu\text{g} / \text{g}$ in Lake Baraki, 3.5 $\mu\text{g} / \text{g}$ in Condonuma and 4.3 $\mu\text{g} / \text{g}$ in Nyu. , 5.1 $\mu\text{g} / \text{g}$ / Akagi Onuma / g, Lake Okutone 7.2 $\mu\text{g} / \text{g}$, Kanako 8.0 $\mu\text{g} / \text{g}$, Lake Kusagi 10 $\mu\text{g} / \text{g}$, Lake Umeda 15 $\mu\text{g} / \text{g}$. The elements having a

correlation coefficient with Cs of 0.8 or more were Ba, Sm, Ce, Eu, and Ta.

2A11 Precise determination of Ir in high purity reagent by neutron activation analysis

MIURA, T.¹, IINUMA, Y.², SEKIMOTO, S.²
(¹NMIJ/AIST., ²KURNS)

Instrumental neutron activation analysis with internal standard correction was applied for precise determine of mg quantities of Ir in high-purity IrBr⁴. A known amount of Au standard solution was used as an internal standard to compensate for inhomogeneity of the neutron flux through an irradiation capsule and for neutron self-shielding. The linearity of the calibration curves of Ir was improved by using an internal standard. The relative expanded uncertainty ($k = 2$) associated with the determined value was 0.86%, which allowed the mg quantities of iridium to be precisely determined.

2B01 Dynamics of polaronic local structures in a colossal-magnetoresistive manganite La_{0.7}Ca_{0.3}MnO₃

SATO, W.¹, KOMATSUDA, S.², SHIMIZU, H.¹, MORIICHI, R.¹, ABE, S.¹, WATANABE, S.¹, KOMATSU, S.¹, TERAJ, T.³, KAWATA, S.⁴, OHKUBO, Y.⁵

(¹Inst. Sci. Eng., Kanazawa Univ., ²Inst. Human Soc. Sci., Kanazawa Univ., ³Grad. Sch. Eng., Osaka Univ., ⁴Dept. Chem. Fukuoka Univ., ⁵Inst. Integ. Radiat. Nucl. Sci., Kyoto Univ.)

The magnetic hyperfine field and electric field gradients at a radioactive impurity probe of ¹¹¹Cd(←^{111m}Cd) occupying the La/Ca *A* site in a colossal-magnetoresistive perovskite manganite La_{0.7}Ca_{0.3}MnO₃ ($T_C \sim 250$ K) were measured by means of time-differential perturbed angular correlation (TDPAC) spectroscopy. In the paramagnetic-insulator phase at room temperature, the ¹¹¹Cd(←^{111m}Cd) probes are distributed in two different environments (distorted and less distorted sites) at the fraction of 7:3; whereas in the ferromagnetic-metal phase below T_C , the oscillatory structure of the distorted

component vanishes from the TDPAC function. The vanishing of the oscillation is ascribable to dynamic motion of polarons dragged by conduction electrons originating from e_g electrons of Mn³⁺. At liquid helium temperature, the dynamic motion freezes, leaving averaged quadrupole frequency reflecting local distortion. The dynamic motion and freezing of local structures associated with the colossal-magnetoresistance phase transition to the ferromagnetic phase is discussed on the basis of temperature-dependent electromagnetic fields at the probe nuclei of the *A* site ions.

2B02 Observation of Dynamic Behavior and Site Occupation of ¹¹¹Cd(←¹¹¹In) Probe Doped in SrTiO₃

KOMATSUDA, S.¹, SATO, W.², OHKUBO, Y.³
(¹Inst. Hum. Soc. Sci., Kanazawa Univ., ²Inst. Sci. Eng., Kanazawa Univ., ³KURNS)

Strontium titanate (SrTiO₃) is a cubic perovskite oxide. SrTiO₃ doped with metal ions as impurities exhibits a wide variety of electronic properties. Especially, SrTiO₃ doped with trivalent metal ions at Ti⁴⁺ site has been attracted much attention because of their excellent photocatalytic activity. For a practical use of SrTiO₃, it is necessary to obtain more microscopic information on the impurity site. Therefore, we investigated the local structures at the In³⁺ site in SrTiO₃ by means of the time-differential perturbed angular correlation (TDPAC) method using the ¹¹¹Cd(←¹¹¹In) probe. The TDPAC spectrum of ¹¹¹Cd(←¹¹¹In) embedded in SrTiO₃ can be reproduced by a fit with three unique quadrupole frequencies. As for one of these components, the quadrupole frequency shows a zero. On the basis of the cubic structure of SrTiO₃, this signifies the ¹¹¹Cd(←¹¹¹In) probes occupy defect-free substitutional Sr or Ti sites in SrTiO₃. The remaining two components shows two well-defined EFG values. These non-zero frequencies imply that defects were associated with ¹¹¹In probes. In the session, based on the further investigation of TDPAC spectra

for $^{111}\text{Cd}(\leftarrow^{111}\text{In})$ embedded in SrTiO_3 measured at various temperature (400~1100 K), we discuss the defect structure of local surroundings of In ions doped in SrTiO_3 .

2B03 Effect of thermal diffusion of In impurities in ZnO on its electrical conductivity

TAKATA, M.¹, SHIMIZU, H.¹, KOMATSUDA, S.², OHKUBO, Y.³, SATO, W.^{1,4}

(¹Grad. Sch. Nat. Sci. Tech., Kanazawa Univ., ²Inst. Human Soc. Sci., Kanazawa Univ., ³Inst. Integ. Radiat. Nucl. Sci., Kyoto Univ., ⁴Inst. Sci. Eng., Kanazawa Univ.)

Zinc oxide (ZnO) is known as an intrinsic n-type semiconductor, and its physical and chemical properties are extensively studied aiming to enhance n-type conductivity by introducing impurity donors. Thus, control of the state of impurities is an urgent issue for its application to functional materials. In our previous study, we succeeded in uniformly diffusing 0.5at.% In ions in ZnO. In the next stage, we directed our interest to thorough understanding of the effect of association-dissociation behavior of impurity In ions on electrical conductivity. In this presentation, the effect of thermal diffusion of In impurities was investigated by means of perturbed angular correlation spectroscopy with the $^{111}\text{Cd}(\leftarrow^{111}\text{In})$ probe and the electrical conductivity measurements.

2B04 Change in gel fraction of polyethylene after irradiation: Evaluation by positron annihilation lifetime spectroscopy

KITADA, N.¹, OKA, T.², OKUTSU, K.¹, YAMASHITA, T.^{3,1}, KINO, Y.¹, SEKINE, T.^{3,1}

(¹Department of Chemistry, Tohoku Univ., ²Japan Atomic Energy Agency, ³Institute for Excellence in Higher Education, Tohoku Univ.)

Polyethylene (PE) is widely used for insulators in nuclear power plants and accelerators. During long-term usage, since PE is subjected to radiation environments, and is degraded by oxidation. The examination of the degradation of PE is important for the safety of these facilities.

In this work, we evaluated the degradation of the gamma-ray irradiated PE with positron annihilation lifetime spectroscopy (PALS). In order to show the effectiveness of the PALS, we compared with gel fraction measurement. PALS is known as a powerful tool to investigate nanostructure without destroying the sample. Ultra-high molecular weight PE (UHMWPE) with a molecular weight of 1 million or more, high-density PE (HDPE) with a molecular weight of about 300,000, and low-density PE (LDPE) having branched molecular chains, were irradiated with gamma-ray. The samples were irradiated under vacuum and room temperature condition and then exposed to the air.

The results of PALS showed that the formation probability of spin-triplet positron-electron bound state, *ortho*-positronium (*o*-Ps), decreased after the irradiation as the air exposure time increased. The gel fraction also decreased as the air exposure time increased. We found a correlation between the gel fraction (0-100% for HDPE and LDPE, and 80-100% for UHMWPE), and the formation probability of *o*-Ps. The relation between the change of the gel fraction and that of the formation probability of *o*-Ps was explained by the oxidation of the sample. The results suggest that PALS can be a nondestructive method for analyzing radiation degradation of polymers as an alternative to gel fraction.

2B05 Observation of oxidation reaction of metal iron by muonic X-ray measurement

KAJINO, M.¹, NINOMIYA, K.¹, KUDO, T.¹, TERADA, K.¹, INAGAKI, M.², SATO, A.¹, TOMONO, D.³, KAWASHIMA, Y.³, SAKAI, Y.⁴, TAKAYAMA, T.⁴, SHINOHARA, A.¹

(¹Osaka Univ., ²Kyoto Univ., ³RCNP, ⁴Daido Univ.)

An atomic system having one negatively charged muon instead of an electron is called a muonic atom. After formation of a muonic atom, high-energy muonic X-rays are emitted in the de-excitation process of the atomic muon. Recently, non-destructive elemental analysis method by muonic X-ray measurement has been

developed. In the formation process of the muonic atom, chemical environmental effects on muonic X-ray intensities have been known. For example, intensity ratio of muonic K_{α} X-ray to K_{β} X-ray is different by chemical species of muon capturing atom. In this work, we performed feasibility study on observing oxidation reaction tracking by muonic X-ray measurement.

Muon irradiation experiments were conducted at RCNP-MuSIC. We prepared a packed iron powder included in disposable heat pad for muon irradiation sample. Muonic X-rays from the sample were continuously measured during its oxidation by HPGe detectors. Muonic K_{α} X-ray intensity ratio from iron became large with time passing. The time dependence on muonic K_{α} X-ray intensity ratio means progress of oxidation process of the sample. Considering the sample as the binary system of iron oxide and metal iron, this time dependence of muonic X-ray intensity was consistent with the chemical species change revealed by Mossbauer spectroscopy. In summary, we successfully demonstrated the chemical reaction tracking by muonic X-ray measurement.

2B06 Development of non-destructive 3D image method using muon beams with CdTe Double-sided Strip Detector.

CHIU, I-H.¹, KAJINO, M.¹, SHINOHARA, A.¹, NINOMIYA, K.¹, TAKEDA, S.², KATSURAGAWA, M.², TOMARU, R.², YABU, G.², NAGASAWA, S.², TAKAHASHI, T.², WATANABE, S.³

(Osaka University¹, Kavli IPMU², JAXA³)

Analyzing the internal elements without destroying the sample is an important target for valuable materials such as the archaeological objects, the meteorites. The non-destructive analysis has been achieved using muon-induced characteristic X-rays with the high-intensity negative muon beam. Since the energy of muon characteristic X-rays is 200 times higher than that of electron characteristic X-rays, it provides a new method to analyze the internal elements and depth of substances by the non-destructive

elemental analysis. Furthermore, a two-dimensional cadmium-telluride detector, which is used for space observation experiments, is possible to be used in the image reproduction with excellent positional resolution. The two-dimensional image has been developing using muon characteristic X-rays so far. A new method for the non-destructive three-dimensional image with muon characteristic X-rays is expected. Moreover, we use the Geant4 program to evaluate the validity of the experimental data. Geant4 program is commonly used in particle physics and medicine and astronomy. In this presentation, we will show the construction of a two-dimensional cadmium telluride detector and the analysis method of the image reproduction. In addition, the non-destructive 3D imaging method using muon characteristic X-rays will be discussed, too.

2B07 Residential Site of Impurity In Atoms in Fe₃O₄ Observed by Means of Positron Annihilation Spectroscopies and Mössbauer Spectroscopy

ISHIZAKI, R.¹, SHIMIZU, H.¹, SATO, W.^{1,2}

(¹Grad. Sch. Nat. Sci. Tech., Kanazawa Univ., ²Inst. Sci. Eng., Kanazawa Univ.)

Iron(II,III) oxide (Fe₃O₄) with an inverse spinel structure has many vacancies at the sites where Fe atoms can occupy; it is expected that enhancement of magnetism and electrical conductivity is attained by introducing impurity ions to the vacant sites. When a trace amount of indium (In) was introduced as impurities in Fe₃O₄ in our previous work, it was found that In occupies the tetrahedral site at room temperature. Our next interest is in the directed to residential sites of a macroscopic amount of impurity In introduced. In this study, we investigated the dependence of In concentration introduced in Fe₃O₄ on the local structure in Fe₃O₄ by means of positron annihilation lifetime spectroscopy (PALS), Doppler broadening spectroscopy (DBS) and ⁵⁷Fe Mössbauer Spectroscopy. PALS and DBS measurements show that In having a large ionic radius substitutes for Fe, squeezing

nearby local structures to cause the reduction of the vacancy volume. The results of Mössbauer measurements revealed that hyperfine fields at the Fe site and the residential fraction of Fe in A and B sites change depending on the In concentration. These observations suggest that In introduced in Fe₃O₄ replaces Fe ions at the A site.

2B08 Mössbauer spectroscopy of Fe-Ag type Hofmann-type complex

KITASE, K., TAKAHASHI, M., KITAZAWA, T. (Toho Univ.)

Hofmann-type complex is a coordination polymer with a center metal ion such as Fe, Ni or Cd, bridging ligand that is liner dicyanide complex or square planer tetracyanide complex, and pyridine-type ligands. The spin-crossover (SCO) phenomenon is a reversible transition between high-spin and low-spin state. ⁵⁷Fe Mössbauer spectroscopy is a useful tool for investigations in Fe-based SCO phenomenon due to dependence of valences, spin states, and environments of Fe site.

In this research, we synthesized a Hofmann-type complex Fe(4-methylpyrimidine)₂[Ag(CN)₂]₂. This complex is synthesized by direct method using Mohr's salt, K[Ag(CN)₂] and ligand. The magnetic measurement shows one-step spin transition between 140 and 130 K in this complex. The single crystal data shows this complex belongs to the Hofmann-type structure. weak Ag — N interaction exists. ⁵⁷Fe Mössbauer spectroscopy of this complex reveals spin transition occurs 140 and 130 K. However, no hysteresis is observed in ⁵⁷Fe Mössbauer data while a relatively large hysteresis is observed in SQUID data. The differences between Mossbauer and SQUID data might be explained by different scan rates. We are set to measure magnetic susceptibility to make sure this explanation.

2B09 Mössbauer spectroscopic studies of Na₂Ru_{1-x}Fe_xO₃ of sodium-ion battery electrode

HAMANO, K.¹, KOBAYASHI, Y.^{1,2}, HABA, H.², UENO, H.²

(¹Univ. Electro-Comm., ²Nishina Center, RIKEN) Sodium-ion battery has attracted attention as the next generation of battery after Li-ion one for large-scale applications owing to the abundance and low cost of sodium. Among Na-excess metal oxides, Na₂RuO₃ is expected to be the leading cathode material. Mössbauer spectroscopy is a powerful tool to understand the oxidation states of Ru ions. However, the measurements of ⁹⁹Ru Mössbauer spectra can only be performed at low temperatures because the Mössbauer transition energy is relatively high ($E\gamma = 89.8$ keV). A ⁵⁷Fe-doped sample of Na₂Ru_{1-x}Fe_xO₃ was prepared to estimate the temperature dependence of the oxidation states of Ru sites by ⁵⁷Fe Mössbauer spectroscopy. In this study, we demonstrate the oxidation states and coordination environments of Ru ions in Na₂RuO₃ and Na₂Ru_{1-x}Fe_xO₃ after repeating the charge/discharge processes by means of ⁹⁹Ru and ⁵⁷Fe Mössbauer spectroscopy, XRD, and electrochemical measurements. The Na₂Ru_{0.99}Fe_{0.01}O₃ sample was confirmed to be in a single phase by XRD. The ⁹⁹Ru Mössbauer spectrum of Na₂Ru_{0.99}Fe_{0.01}O₃ after charging showed that Ru ions were oxidized from Ru⁴⁺ before charging to Ru⁵⁺. We will discuss that the valence states and structural changes of Ru ions due to charging-induced Na⁺ deficiencies from the results of ⁵⁷Fe Mössbauer spectroscopy.

2B10 In-beam Mössbauer spectra of ⁵⁷Fe after ⁵⁷Mn implantation in alkali halides

KIJI, M.¹, HAMANO, K.¹, SATO, M.¹, TAKAHAMA, N.¹, KOBAYASHI, Y.^{1,2}, KUBO, M.K.³, YAMADA, Y.⁴, MIHARA, M.⁵, SATO, W.⁶, NAGATOMO, T.², OKAZAWA, A.⁷, SATO, S.⁸, KITAGAWA, A.⁸

(¹Univ. Electro-Commun., ²RIKEN Nishina Center, ³Int. Christ. Univ., ⁴Tokyo Univ. Sci., ⁵Osaka Univ., ⁶Kanazawa Univ., ⁷Nihon Univ., ⁸National Institute of Radiological Science)

In-beam Mössbauer spectroscopy using a short-lived ⁵⁷Mn ($T_{1/2} = 1.45$ m) implantation is a powerful tool to study the characterization of localized ⁵⁷Fe atoms in solids. Alkali halide is

mainly used as an optical material. Since iron is mixed as an impurity in the optical material during the manufacturing process, there is a problem that the visible light transmittance becomes lowered. In order to use an optical material with high efficiency, it is necessary to have high purity. However, it is very difficult to quantify a trace amount of impurities, and it is not known how it behaves. Therefore, we conducted a study for understanding the occupied position and chemical state of ^{57}Fe in alkali halides by using in-beam Mössbauer spectroscopy and DFT calculation. All the obtained Mössbauer spectra could be analyzed with three components, one singlet and two doublets, respectively. From the obtained Mössbauer parameters and DFT calculations, the singlet and doublets were assigned to the substituting positions of the cations without and with defects, respectively. In addition, we will discuss the characteristic behavior of Fe atoms in LiF, NaF, NaCl, and KCl, which have NaCl-type crystal structures.

2B11 Identification of Co sites in spinel ferrite by means of ^{57}Co emission Mössbauer spectroscopy

SAKAGUCHI, M.¹, TAKENAKA, S.¹, SATO, W.^{1,2}

(¹Grad. Sch. Nat. Sci. Tech., Kanazawa Univ.,
²Inst. Sci. Eng., Kanazawa Univ.)

Cobalt ferrite ($\text{Co}_x\text{Fe}_{3-x}\text{O}_4$) is a magnetic material that has been extensively studied for a wide application in various fields of industry. In spite of the expectation to application, however, even cation distribution is still unknown. Thus, we prepared ^{57}Co -doped CoFe_2O_4 and applied emission and transmission Mössbauer spectroscopy with the $^{57}\text{Fe}(\leftarrow ^{57}\text{Co})$ probe to clarify how cations are distributed in the compound with a spinel structure. It was found that the ^{57}Fe transmission and ^{57}Co emission spectra of CoFe_2O_4 obtained at 7-295 K consist of three components: two components are assigned to the B site and the other to the A site. This result indicates that part of the A sites are occupied by Co ions. On the basis of a model

of probability distribution, it was estimated that $7.5 \pm 0.5\%$ of the A sites are occupied by Co ions. In the session, temperature variation of the spectra are discussed in relation to the ^{57}Co emission Mössbauer spectra for Fe_3O_4 based on the Mössbauer parameters obtained.

3A01 Production and chemical purification of Ag-111 toward nuclear medical application

OOE, K.^{1,2}, WATABE, T.^{1,2}, SHIRAKAMI, Y.², MORI, D.³, YOKOKITA, T.³, KOMORI, Y.³, HABA, H.³, HATAZAWA, J.^{2,4}

(¹Osaka Univ. Grad. Med., ²IRS, Osaka Univ.,
³RIKEN, ⁴RCNP, Osaka Univ.)

Toward domestic production of therapeutic radionuclides using accelerators, production cross sections and chemical purification method of a β -particle emitter, ^{111}Ag , were investigated. A stack of metallic ^{nat}Pd target foils was irradiated with a 24-MeV deuteron beam from the RIKEN K70 AVF Cyclotron. The irradiated ^{nat}Pd targets were subjected to γ -ray spectrometry with a Ge detector. The 245-keV γ peak of ^{111}Ag was used for determination of the cross section of the $^{nat}\text{Pd}(d,x)^{111}\text{Ag}$ reactions. In the chemical separation of ^{111}Ag from the ^{nat}Pd target, the irradiated ^{nat}Pd target was dissolved in mixed HNO_3 and HCl solution. After evaporation to dryness, the residue was dissolved in 1 M HNO_3 solution and then fed into an anion exchange column. The ^{111}Ag was eluted by 1 M HNO_3 solution. The maximum cross section of ^{111}Ag was approximately 40 mb at the deuteron beam energy of approximately 9 MeV. The elution of ^{111}Ag from an anion exchange column using 1 M HNO_3 was completed before the start of elution of ^{nat}Pd . The recovery of ^{111}Ag was more than 99%. The production and separation methods of ^{111}Ag from the metallic Pd were established. These results lead to preclinical studies of therapeutic effects with ^{111}Ag .

3A02 Development of new diagnostic method by ETCC imaging using Tc-95 and Tc-95

HATSUKAWA, Y.¹, KABUKI, S.², KUSHIDA, J.³, IKUTA, A.³, KIMURA, H.⁴, ASAI, M.⁵, TSUKADA, K.⁵

(¹QST, ²Tokai Univ., Medical school, ³Tokai Univ., Physics, ⁴ Kyoto Pharmaceutical Univ., ⁵JAEA)

In this study, we produced Tc isotopes with high energy gamma emitting for developing of Compton camera imaging instead of SPECT measurement with ^{99m}Tc. Technetium-99m is used in radioactive diagnostic tests, for example as a radioactive tracer that a nuclear medical gamma camera can detect in human body. There are at least 31 commonly used radiopharmaceuticals based on ^{99m}Tc for imaging and functional studies of the brain, myocardium, thyroid, lungs, liver, gallbladder, kidneys, skeleton, blood, and tumors. Two technetium isotopes, ^{95m}Tc and ⁹⁶Tc are candidates for Compton camera imaging. The feasibility study of Compton camera imaging using gamma rays emitted from ⁹⁶Tc was carried out. Using the ETCC (Electron Tracking Compton Camera), we tried to obtain imaging pictures of the gamma rays emitted from ⁹⁶Tc. ⁹⁶Tc was obtained from proton irradiation. After chemical purification, ⁹⁶TcO₄⁻ ions were labeled with DTPA compound. Then the ⁹⁶Tc labelled DTPA compound were injected into a mouse, and imaging picture of the mouse was taken by the ETCC imaging method. Detailed results will be discussed in the conference.

3A03 Imaging the arsenic translocation and accumulation in arsenic hyperaccumulator *Pteris vittata* using Positron Emitting Tracer Imaging System(PETIS)

KOHDA, T.¹, QIAN, Z.J.¹, CHIEN, M.F.¹, INOUE, C.¹, IKEDA, H.^{2,3}, WATABE, H.², KIKUNAGA, H.³, SUZUI, N.⁴, YIN, Y.G.⁴, KAWACHI, N.⁴, KITAJIMA, N.⁵

(¹GSES, Tohoku Univ., ²CYRIC, Tohoku Univ., ³ELPH, Tohoku Univ., ⁴QST, ⁵TDD, Fujita Corp.)

Arsenic (As) contamination of drinking water and irrigation water became a serious issue to

millions of people in the world. As remediation in water employs granular metal oxide adsorption method, ion exchange, or coagulation–microfiltration method. The main disadvantage of these methods is high cost of the arsenic removal. Phytoremediation is an emerging water treatment technology that uses the plants to remove contaminants from water. *Pteris vittata*, the As-hyperaccumulator plant, which the capability is accumulating large amount of As into fronds and rhizomes. The mechanism of As accumulation in this plant is still keeping unknown. In the present work, we used PETIS (positron-emitting tracer imaging system) to get the serial noninvasive images and quantitative characterization of absorption, translocation, and accumulation of arsenic (As) in *Pteris vittata* with positron-emitting ⁷⁴As-labeled arsenate tracer. We first recorded the real-time As translocation from the medium to the roots and then to the rhizomes and fronds in *Pteris vittata*. We found that not like roots, rhizomes keep the As level with fronds indicates that rhizomes control the As translocation to fronds in *Pteris vittata*. But non-accumulator *Arabidopsis thaliana* hold most of As in roots.

3A04 Protective behavior of tea catechins on DNA double strand breaks by radiation

WADA, T.¹, YAMAZAKI, S.¹, KOIKE, A.¹, ASHIZAWA, K.¹, ZHAO, M.¹, SUN, F.¹, OYA, Y.²

(¹Graduate School of Integrated Science and Technology, Shizuoka Univ., ²Faculty of Science, Shizuoka Univ.)

The evaluation of radiation effects for human body is necessary. Regarding on a safety issue on radiation damages for human being, it is important to elucidate the protecting mechanism for natural antioxidants including vitamin E, caffeine and catechin from radiations. In the present study, DNA double strand breaks on genome-sized DNA was evaluated quantitatively under several types of radiation sources and the characteristics of radiation damages in different LET was revealed, and the effect of radiation

protection by tea catechins was also studied as a function of its concentration. The DNA of bacteriophage (T4 GT7: 166 kbp) was soaked in a sterilized water with EGCg ((-)-epigallocatechin gallate) which is the main ingredient of a tea catechin and exposed to various radiation sources (^3H - β ray, ^{45}Ca - β ray, ^{60}Co - γ ray). The length of DNA molecules was measured using fluorescence microscope, which can estimate a number of DSBs quantitatively. Based on experimental results, the DSBs were reduced by addition of EGCg and a saturation on the protection in EGCg concentration of 1 μM was found. As it was reported in previous study that the concentration of EGCg in blood was increased to several μM after ingesting tea extract, the result suggested that EGCg could protect organisms from indirect radiation effect.

3A05 Search for an optimized condition of ^{207}Po elimination for the $^{211}\text{Rn}/^{211}\text{At}$ generator system

AOI, K.¹, SHIN, Y.¹, KAWASAKI, K.¹, MARUYAMA, S.¹, WASHIYAMA, K.², NISHINAKA, I.³, HABA, H.⁴, WANG, Y.⁴, YOKOYAMA, A.¹

(¹Kanazawa Univ., ²Fukushima Medical Univ., ³QST, ⁴RIKEN)

The astatine-211 nuclide is a prospective candidate to utilize in targeted alpha therapy. To extend usable time of ^{211}At of 7.2h half-life, the $^{211}\text{Rn} / ^{211}\text{At}$ generator system has been in developing. However, ^{207}Po ($t_{1/2} = 5.80$ h) from the decay of ^{211}Rn is also produced as a by-product. It is necessary to separate ^{211}At from ^{207}Po , since ^{207}Po may damage normal tissue of human body. In this study, an optimal condition for elimination of ^{207}Po was studied by a cation exchange resin of 0.1, 0.3, 0.5, and 0.7 g, and ascorbic acid of 0.1, 0.5, 1, 2, and 3wt%, in 0.01M HCl as eluent. The results demonstrate the absorption rate of more than 90% of ^{207}Po was obtained for cation exchange resin. However, the recovery rate of about 94% of ^{211}At from cation exchange resin of 0.5 g in 0.01M HCl was most high for ascorbic acid of 1wt%. As the weight of

resin increased, the recovery rate of ^{211}At increased, and the absorption rate of ^{207}Po decreased. Separation factor was 35 for cation exchange resin of 0.5 g with 1wt% ascorbic acid in 0.01M HCl. Thus, this is a suitable condition for the elimination method of ^{207}Po from ^{211}At .

3A06 Study on solvent extraction and speciation of astatine species with a thin layer chromatography

MARUYAMA, S.¹, KAWASAKI, K.¹, AOI, K.¹, HIGASHI, M.², NISHINAKA, I.², WASHIYAMA, K.³, HABA, H.⁴, YOKOYAMA, A.⁵

(¹Kanazawa Univ., ²QST, ³Fukushima Med. Univ., ⁴RIKEN)

In recent years, studies on α -ray internal therapy using α -ray emitting nuclides have been actively conducted, and astatine-211(^{211}At) has been attracting much attention from the viewpoints of availability of nuclides and outpatient treatment. Astatine is a radioactive element that has only short-lived radionuclides, and various chemical species of At have been reported in relation to its chemical behaviors that were not well clarified. We obtained the nuclide from the $^{209}\text{Bi}(\alpha, 2n)^{211}\text{At}$ reaction to study the distribution behavior in solvent extraction in relation to At species. We performed solvent extraction in HCl/DIPE system and speciation of At species with a thin layer chromatography (TLC) followed by Image Plate (IP) measurement on its radioactivity to identify the chemical species of At in a solution and to determine its distribution.

3A07 Synthesis of astatin-211 labeled gold nanoparticles and evaluation of anti-tumor effect

HUANG, X.¹, KATO, H.², KADONAGA, Y.³, SHIMOYAMA, A.¹, KABAYAMA, K.¹, OOE, K.², TOYOSHIMA, A.³, SHINOHARA, A.¹, FUKASE, K.¹

(¹Graduate School of Science, Osaka Univ., ²Graduate School of Medicine, Osaka Univ., ³Institute for Radiation Sciences, Osaka Univ.)

Much attention has been drawn to the α -emission radiotherapy with regard to its potential cancer therapeutic efficacy. Astatin-211 (^{211}At), with a comparatively short half-life of 7.2 h, is able to emit alpha particles within a few cell diameters with high energy, which can damage cancer cells in a relatively high biological effectiveness. ^{211}At has similar characteristics to the iodine, which can rapidly diffuse throughout the body and accumulate in thyroid gland. Since ^{211}At has high affinity to gold, we planned to develop a cancer treatment by direct injection of ^{211}At -labeled gold nanoparticles (AuNP) to cancer cells. Here, we synthesized different sizes of poly(ethylene glycol) methyl ether thiol (mPEG-SH) modified AuNP and performed astatination (^{211}At) of pegylated AuNP (AuNP-S-mPEG). Furthermore, anti-tumor effect of the ^{211}At -AuNP-S-mPEG were evaluated by using rat xenograft model (C6 glioma). In the results, all 13 nm-120 nm of AuNP-S-mPEG were quantitatively assassinated by simple mixing with ^{211}At solution. *In vivo* studies revealed that the 30 nm ^{211}At -AuNP-S-mPEG was diffused more efficiently in tumor tissue without leaking to blood vessel and has significant anti-tumor effect, whereas the effect of 120 nm size was weaker. The results may help to optimize the design of targeted alpha therapy drugs, which are suitable for intratumorally injection.

3A08 Analytical method for radioactivity and chemical forms of ^{211}At

SEGAWA, M.¹, MAEDA, M.¹, TOH, Y.¹, NISHINAKA, I.², WATANABE, S.², ISHIOKA, N.²

(¹Japan Atomic Energy Agency, ²National Institute for Quantum and Radiological Science and Technology)

^{211}At is artificially produced for medical purposes. To prevent severe loss of short-lived ^{211}At , a rapid method analyzing both the radioactivity and all chemical forms was required for related researches. We propose a new method by means of an α -scintillation-camera system comprising of thin-layer chromatography and a

high-sensitive CCD camera. Our previous research verified the performance of the system for low-radioactivity ^{211}At experimentally. In this study, the performance of the system and its capability for high radioactive ^{211}At are reported.

3A09 Preparation of Pure Sodium Astatide, [^{211}At]NaAt, for Nuclear Medicine Application

SHIRAKAMI, Y., OOE, K., KANEDA, K., TERAMOTO, T., WATABE, T., TOYOSHIMA, A., SHINOHARA, A., HATAZAWA, J.

(Osaka Univ.)

Sodium astatide, [^{211}At]NaAt, is expected to be a good agent for the treatment of patients with thyroid cancer, although it has not yet been used in clinics. We tried to elucidated chemical properties of the aqueous solution of ^{211}At after addition of ascorbic acid.

^{211}At was produced through the $^{209}\text{Bi}(\alpha,2n)^{211}\text{At}$ reaction. Dry-distilled ^{211}At was collected and dissolved in distilled water (^{211}At bulk solution). Into the bulk solution, ascorbic acid and sodium hydrogen carbonate were added and stand for 1hr at room temperature. The solutions were analyzed by thin layer chromatography (TLC) and high performance liquid chromatography (HPLC).

TLC profiles suggested that the ^{211}At bulk solution composed of several ^{211}At labeled species which might be attributable to compounds of At[I] and At[III] oxidation states. After addition of ascorbic acid in the bulk solution, the radioactive species were converged on a single compound. The compound was estimated to be [^{211}At]-NaAt.

We demonstrated a method for the preparation of pure sodium astatide, [^{211}At]NaAt, for the treatment of patients with thyroid cancer.

3A10 Investigation of drug for ^{211}At -labeled nuclear medicine targeting to cancer specific amino acid transporter

KANEDA-NAKASHIMA, K.^{1,2}, SHIRAKAMI, Y.^{1,3}, OOE, K.^{1,4}, TERAMOTO, T.¹, WATABE, T.^{1,4}, SHIMOYAMA, A.^{1,2}, MANABE, Y.^{1,2},

KABAYAMA, K.^{1,2}, TOYOSHIMA, A.^{1,2,3},
FUKASE, K.^{1,2}, SHINOHARA, A.^{1,2}
(¹Osaka Univ., ²Toyonaka College)

Targeted alpha therapy (TAT) is receiving much attention in the field of “theranostics” because of its high cytotoxic effect to the targeting cancer. However, physiological uptakes in non-targeted organs are also observed in the TAT, which might lead to the severe side effects. We should consider about both maximizing the treatment effect in the tumor and minimizing the side effects in the organs at risk. From this viewpoint, decision of targeting molecule was most important.

We selected LAT1 as molecular target. LAT1 is one of the amino acid transporters, which has highly specificity to cancer tissues. We developed next-generation internal radiotherapy using chemicals targeting LAT1. First, we synthesized large neutral amino acid derivative labeled with ²¹¹At. ²¹¹At was produced by the cyclotron, and then quickly purified and labeled. Next, we performed its cytotoxicity evaluation using some cancer cell lines. As a result, cell death and specificity were confirmed. We also confirmed the specificity *in vivo* and found the anti-cancer effects *in vivo* study. In the immediate future, we will examine that the effects of ²¹¹At-labeled amino acid derivative using several kinds of tumor-bearing models.

3A11 Preparation and stability evaluation of α -emitter ²¹¹At-labeled iminobiotin

WASHIYAMA, K., AOKI, M., TATSUMI, T.,
YAMATSUGU, K., SUGIYAMA, A.,
NISHIJIMA, K., JOHO, T., GAO, H., ZHAO, S.,
KANAI, M., KODAMA, T., TAKAHASHI, K.
(¹Advanced Clin. Res Center, Fukushima Med.
Univ., ²Grad. Sch. Pharm. Sci., Univ. Tokyo, ³Isot.
Sci. Center, Univ. Tokyo, ⁴ Res. Center Advanced
Sci. Tech., Univ. Tokyo)

By separating the role of antibody and radioligand, pretargeting radioimmunotherapy increases the absorbed dose to the tumor while reducing the side effects on normal tissues. We have developed a pretargeting system using a

mutated low immunogenic streptavidin called as Cupid, which can be fused to four scFv of mAb, and a bis-iminobiotin compound Psyche that specifically binds to Cupid. The alpha emitter, ²¹¹At, is expected to be loaded into Psyche as it irradiates tumors with high-energy radiation with a short-range. The purpose of this study was to optimize the labeling of ²¹¹At to several Psyche compounds (Psyche-B, -J, -N, and -P) and evaluate its stability in PBS and mouse plasma. Organotin precursors of Psyches which modified its structure on Cupid binding site, were synthesized. ²¹¹At was produced using MP-30 cyclotron at Fukushima Medical University and separated by dry distillation using a quartz tube furnace. Psyche precursors were labeled with ²¹¹At using N-bromosuccinimide, and the stabilities in PBS and murine plasma were studied.

²¹¹At-Psyches obtained with 65-96% radiochemical yields and >95% radiochemical purities. All ²¹¹At-Psyches showed high stability in PBS. While ²¹¹At-Psyche-N and -P are relatively stable in plasma, ²¹¹At-Psyche-B and -J dissociated in plasma within 1 hr.

3B01 Transport of Soil Particle in the Reservoir-Catchment System Using Fallout Radionuclides ⁷Be and ²¹⁰Pb

TSURUOKA, M.¹, OCHIAI, S.², MATSUKI, A.²,
NAGAO, S.²

(¹Graduate School of Natural Science and
Technology, Kanazawa University., ²Institute of
Nature and Environmental Technology,
Kanazawa University)

In this study, transport of surface soil particle in the reservoir-catchment system located in Noto Peninsula, was investigated by using fallout radionuclides ⁷Be (half-life 53 days) and ²¹⁰Pb (half-life 22 years). The amount of radionuclides depositing from the atmosphere, sedimentation in reservoir, and the outflow from system were measured using a collecting pan, sediment trap, and water level meter installed in the reservoir. The amount of radionuclides transported from the catchment area to the

reservoir was estimated assuming that the total amount of deposition from the atmosphere and transport from the catchment corresponded to the total amount of sedimentation in reservoir and outflow from the system. The amount of ^{210}Pb accumulated in the reservoir was mostly due to the amount transported from the catchment area. On the other hand, the amount of ^7Be transported from the catchment area was smaller than that of ^{210}Pb . It suggests that time scale of soil particle transportation is much longer than the half-life of ^7Be in this catchment. This result may be attributed to the result that most of the ^7Be is trapped in the litter layer in catchment soils implying the slow migration of ^7Be from litter to soil particles.

3B02 Uranium reduction by Fe(II) in smectite TAKEDA, S.¹, SUN, J.¹, HAMAMOTO, T.², ISHIDA, K.², TANAKA, M.¹, ITAI, T.¹, TAKAHASHI, Y.¹

(¹The Univ. of Tokyo, ²NUMO)

Redox behavior in underground geosphere is important in terms of radioactive-waste management and enrichment of uranium to form uranium deposit. We conducted an experiment to study if uranium (VI) is reduced into uranium (IV) by Fe(II) in smectite. Sodium dithionite was used to reduce Fe^{3+} in Kunipia F, $(\text{Na}_{0.42}\text{K}_{0.008}\text{Ca}_{0.068})(\text{Si}_{13.91}\text{Al}_{0.09})(\text{Al}_{1.56}\text{Mg}_{0.31}\text{Fe}^{3+}_{0.09}\text{Fe}^{2+}_{0.01})\text{O}_{10}(\text{OH})_2$. Uranyl solution was added to the suspension of Fe(II)-bearing smectite as an adsorbent. All the procedures were conducted in a glove box to avoid oxidation. XANES analysis showed that U(IV)/U(VI) ratio increased as the increase of Fe(II)/Fe(III) within the octahedral layer in the Kunipia F, whereas almost all uranium adsorbed to Kunipia F was U(VI) after its contact with Fe(III)-bearing Kunipia F without the reduction treatment. In the presence of carbonate ions in the solution, U(IV) was stabilized as carbonate complex, which is less likely to be reduced. Bond length of Fe(II)-O in the smectite was smaller than that of Fe(II)-O in regular Fe octahedral compounds, which may

lead to the tendency for Fe^{2+} to be oxidized to Fe^{3+} .

3B03 Fixation of radium by adsorption on clay minerals in the environment

YAMAGUCHI, A.^{1,2}, KURIHARA, Y.¹,
OKUMURA, M.¹, HIGAKI, S.², TAKAHASHI,
Y.²

(¹JAEA, ²Univ. of Tokyo)

Radium (Ra), which is generated by radioactive decay of uranium and thorium, is an important element from the standpoints of radiation protection and groundwater circulation. Although some previous studies suggested the fixation of Ra by clay minerals based on relatively higher adsorption coefficients, its detail has been unclear. Therefore, the purpose in this study is to investigate the migration of Ra in the environment with clay minerals. In this study, core samples collected around Ningyo-toge uranium mine were analyzed because they had high concentrations of Ra and large amounts of clay minerals in weathered granite layer. The samples were analyzed by many methods, including γ -ray spectroscopy, sequential extraction, and X-ray absorption fine structure (XAFS). The activity ratio of $^{226}\text{Ra}/^{238}\text{U}$ was below and over 1 in the concentrated layer and the weathered granite layer, respectively. Results of sequential extraction and XAFS showed that more amount of Ra was distributed to clay minerals. These results suggested that Ra is adsorbed on clay minerals and fixed in weathered granite layers, which is important not only for clarification of the behavior, but for development of removal method of Ra.

3B04 Nationwide survey of atmospheric ^{85}Kr activity concentration in Japan

SATO, Y., NITTA, W., ISOGAI, K.

(Japan Chemical Analysis Center)

^{85}Kr (half-life: 10.76 year) is a major radio nuclide released from the spent nuclear fuel reprocessing plants into the atmosphere. In Japan, a large-scale nuclear fuel reprocessing plant will be operated in Rokkasho. Prior to the beginning

of full operation, a nationwide survey of atmospheric ^{85}Kr has been carried out since 2006. Atmospheric ^{85}Kr concentrations from 2006 to 2019 were within the range of 1.30 to 1.60 Bq / m^3 (Average: 1.46 Bq / m^3) expect for the temporary increases due to the pilot operation of domestic reprocessing plants and the accident of Fukushima Daiichi Nuclear Power Plant.

Considering the global inventory of ^{85}Kr and the annual decreasing rate due to radioactive decay, It can be presumed that about 340 PBq (P(peta)= 10^{15}) of ^{85}Kr are removed from the atmosphere each year. From this presumption and the annual release of ^{85}Kr from reprocessing plants (La Hague, Sellafield, Rokkasho), it is possible to estimate whether annual ^{85}Kr inventory increase or decrease. Comparing the changes in ^{85}Kr inventory with the ^{85}Kr concentrations in Chiba since 2006 obtained in this survey, it was recognized that there was a relationship with the operating status of reprocessing plants around the world.

3B05 Spatial distributions of $^{234}\text{Th}/^{238}\text{U}$ activity ratio in the Japan Sea and the implications for scavenging effects

KAMEYAMA, H.¹, INOUE, M.¹, HANAKI, S.¹, MOROKADO, T.¹, SHIROTANI, Y.¹, TAKEHARA, R.¹, MORITA, T.², MIKI, S.², HONDA, N.³, NAGAO, S.¹

(¹Kanazawa University, ²National Research Institute of Fisheries Science, ³Japan Sea National Fisheries Research Institute)

Reactive (particle-reactive) radionuclides in the seawater have been utilized to estimate the scavenging from seawater. The activities of the reactive ^{234}Th (half-life, 24 d) and the extent of disequilibrium with its soluble parent ^{238}U ($^{234}\text{Th}/^{238}\text{U}$ activity ratio) in seawater clarified the scavenging processes. To study the circulation of particles in the Sea of Japan, water samples were collected from 0-1000 m depths in the Tsushima Warm Current area of the Japan Sea in July-August of 2016-2019 and February of 2019-2020. By employing low-background γ -spectrometry, the activities of ^{228}Th (half-life,

1.91 y) and ^{234}Th were determined along with ^{226}Ra (half-life, 1600 y) and ^{228}Ra (half-life, 5.75 y). In the present study, we examined the vertical and seasonal profiles of $^{228}\text{Th}/^{228}\text{Ra}$ ratio (and $^{228}\text{Th}/^{226}\text{Ra}$ ratio) in the Japan Sea. Combining with water circulations implied from $^{228}\text{Ra}/^{226}\text{Ra}$ ratio, we clarify vertical behaviors of reactive materials in the Sea of Japan.

3B06 Spatial distribution of ^{134}Cs concentration in the surface waters of the northwestern North Pacific Ocean

TAKEHARA, R.¹, INOUE, M.¹, HANAKI, S.¹, MATSUNAKA, T.¹, MORITA, T.², MIKI, S.², KURODA, H.², TANIUCHI, Y.², KASAI, H.², NAGAO, S.¹

(¹Kanazawa University, ²National Research Institute of Fisheries Science)

This study examined the spatial profiles of radiocesium (^{134}Cs and ^{137}Cs) concentrations (activities) in the surface waters of the northwestern North Pacific Ocean in 2018-2019. Because of its short half-life (2.06 y), the ^{134}Cs detected in seawater samples in the present study is believed to be entirely derived from the Dai-ichi Nuclear Power plant (FDNPP) accident. Therefore, to investigate the migration patterns of FDNPP-derived radiocesium after the accident, we mainly focus on ^{134}Cs concentrations on the FDNPP accident date (March 11, 2011). To measure low-level ^{134}Cs concentrations, low-background gamma-spectrometry was performed on all seawater samples using Ge-detectors at the Ogoya Underground Laboratory, Japan. In this study, we mainly focus on the offshore of the eastern Hokkaido, where the water masses (Oyashio Current, Soya Warm Current, and surface water of the Sea of Okhotsk) are complicatedly mixed. We classify the pattern of the water mass mixing, using the data of ^{134}Cs combining with Radium isotope and salinity.

3B07 Adsorption behavior of cations in seawater on barium silicate-based adsorbent for Sr

MINOWA, H.¹, OGATA, Y.², KATO, Y.³, KOJIMA, S.⁴

(¹Jikei Univ., ²Nagoya Univ., ³Hitachi Ltd., ⁴Aichi Med. Univ.)

Sr adsorbent (Pureceram MAq®, Nippon Kagaku Kogyo) which based barium silicate (BaSi₂O₅) is useful for analyzing radiocesium in the environment because it specifically adsorbs Sr. The purpose of this study is to explain the behavior of elements such as Sr, Cs, Na, K, Mg, Ca, Ra and Pb in the system of liquid phase: artificial seawater and solid phase: Sr adsorbent. We report the partition coefficients of this adsorbent and seawater for these elements. The liquid phase residual ratio of Cs, Na, K, Mg, Ca is 99% or more, and these elements are distributed in the liquid phase. Therefore, Sr can be easily separated from these elements. The residual rate of Pb in the liquid phase is 14%, and Pb remains in the liquid phase. On the other hand, Ra and Ac adsorb quantitatively to the solid phase like Sr. It has been suggested that SrSO₄, which is insoluble, is involved in the adsorption of Sr in this adsorbent. Ra and Pb are also adsorbed in the state of sulfates as RaSO₄ and PbSO₄. It was suggested that this adsorbent could be used not only for Sr but also for collecting Ra and Ac from seawater.

3B08 Development of analytical method of ¹³⁵Cs/¹³⁷Cs isotope ratio in environmental soil samples

SHIMADA, A.¹, KIM, M.², TSUKAHARA, T.², NOMURA, M.², SHIMADA, T.¹, TAKEDA, S.¹, YAMAGUCHI, T.¹

(¹Japan Atomic Energy Agency, ²Tokyo Institute of Technology)

It is possible to deduce contribution of the Fukushima Daiichi Nuclear Power Station (1F) accident from the detected radioactivity of ¹³⁷Cs as background at the completion of decommissioning of nuclear facilities. In order to identify origin of ¹³⁷Cs using ¹³⁵Cs/¹³⁷Cs isotope ratio, radiocesium must be separated from major elements of soil followed by measurement of mass spectrometry. In this study, Cs separation

method using Calix[4]arene-bis(tertoctylbenzo-crown-6) (BOBCalixC6) was developed.

First, reusability of the BOBCalixC6 in 1-octanol solution was studied. The extraction, stripping, and back-extraction properties for Na, Mg, Al, K, Ca, Fe, Sr, Mo, Sn, Sb, Cs, and Ba were hardly changed by 5 times usage and Cs was separated from the other elements. Therefore, approximately 150 ml of the dissolved solution of soil sample collected at Tokai-mura was treated with 2 sets of 15 ml of BOBCalixC6 solution 5 times. The ¹³⁵Cs/¹³⁷Cs isotope ratio was determined using thermal ionization mass spectrometry. Although precise value of 0.3634±0.0005 was obtained for 16900 Bq/g soil collected near 1F, the precision was decreased for low concentration of soil samples collected in Tokai-mura.

3B09 Spectrum analysis of low energy radiation using a plastic scintillator bottle

KATO, Y.¹, OGATA, Y.², MINOWA, H.³, KOJIMA, S.⁴

(¹Hitachi, Ltd., ²Nagoya Univ., ³Jikei Univ., ⁴Aichi Med. Univ.)

We have researched and presented for the simple and rapid analysis of radiocesium in seawater. In this work the plastic scintillator bottle (PSB) method was developed. The PSB method uses a plastic scintillator to measure a filter sample with a low background liquid scintillation counter (LSC-LB7, Hitachi). One of the features of this method is to pouch the sample to prevent contamination. We have succeeded in improving the PSB method by examining the material for the pouch processing, and it has become possible to measure β-ray emitting nuclides with high counting efficiency. We thought that this measurement method could be applied to other nuclides. Therefore, typical low energy β-emitting nuclides, ³H, ⁶³Ni and ¹⁴C, were measured and spectrally analyzed. By using the film of plastic scintillator for the new pouch processing, it was possible to obtain counting

efficiencies of 14.4%, 37.0% and 83.7% for ^3H , ^{63}Ni and ^{14}C , respectively.

3B10 Educational use of Ge-68/Ga-68 generator under legally defined level as radioactive material (3): introduction to experimental program on role of humics in soil environment

MINAI, Y.^{1,2}, NIIZAWA, K.³, NAGATSU, K.⁴, WASHIYAMA, K.⁵, NOZAKI, T.^{3,6}

(¹Musashi Univ., ²Nedzu Chem. Lab., ³Kitasato Univ., ⁴NIRS, ⁵Fukushima Med. Univ., ⁶RIKEN) Radionuclides under particular level of radioactivity is not regarded as radioisotopes under current regulations. We can apply very trace radioactivity for educational purposes at classrooms without special facility for handling of radioactive substances. Our group supporting Dr. T. Nozaki has focused on development of $^{68}\text{Ge}/^{68}\text{Ga}$ generator kit under its regulation level, 100kBq, for education.

This is the third report after publication of the summary of achievements made at the initial stage of development in 2018. This report proposes a use of the $^{68}\text{Ge}/^{68}\text{Ga}$ generator kit for education. A laboratory research program in environmental science for high school students was chosen as a trial case for developing a new education program using the generator. The program was focused on experimental works to understand roles of humics in soil environment. The program composed of 9 modules of experiments includes 2 modules on metal-humate interaction: adsorption of metal cations on humic precipitates and metal-humate complex formation. Non-radioactive Cu^{2+} or Co^{2+} was used in those modules. Color of each metal ion is an indicator of the presence of the ion in solution contacting with humic precipitates or other solid media. This report proposes that the ^{68}Ga tracer can replace those visible cations applied in a laboratory research program. Besides, it is possible to improve quality of experiments, using the tracer.

3B11 Radiocarbon dating of Bifukumon'in-Fujiwara-no-Nariko-hotsugan-konshi-kinji-issaikyo sutra: Natural scientific evidence for the use of Zn in the Heian period, Japan

ODA, H.¹, NISHIYAMA, Y.², IKEDA, K.³

(¹Nagoya Univ., ²Nara Univ., ³Chuo Univ.)

Bifukumon'in-Fujiwara-no-Nariko-hotsugan-konshi-kinji-issaikyo sutra has been considered to have been written on navy blue paper with gold mud. Recent studies, however, indicated that the gold letters are by brass (Cu and Zn), not by Au. To clarify whether the sutra was truly written in the Heian period, and whether Zn was used in Japan at that time, we measured radiocarbon age of the sutra. Paper sample weighting ca.10mg were cut from the margin of the calligraphy. The sample was soaked in H_2O to peel away the original sheet from the lining. The original sheet and the lining were then washed in H_2O , 1.2N HCl and 1.2N NaOH. Graphite target was prepared from each sample. Radiocarbon ages were measured using the CAMS-500(NEC, U.S.A) at Paleo Labo Co., Ltd., Japan. The calibrated radiocarbon age of the original sheet indicated 1020-1151 cal AD. It shows that the sutra was indeed written in the Heian period and brass was used during this period.

P01 Solvent extraction of Fr and Cs with calix[4]arene-bis(benzocrown-6)

KOMORI, Y., HABA, H.

(RIKEN Nishina Center)

Francium (Fr) is the heaviest alkali metal. Fr has been one of the least-studied elements among the naturally occurring elements because it has no stable isotopes and all its isotopes are short-lived. Due to the experimental difficulties, chemical properties of Fr have not been studied in detail so far. In this work, we investigated the solvent extraction behavior of Fr and Cs using calix[4]arene-bis(benzocrown-6). 700 μL of 10^{-4} – 3 M NaNO_3 aqueous solutions containing ^{221}Fr milked from an $^{225}\text{Ac}/^{221}\text{Fr}$ generator and ^{137}Cs were transferred into a polypropylene tube. 700 μL of $1.5 \times 10^{-3} \text{ M}$

calix[4]arene-bis(benzocrown-6) in 1,2-dichloroethane was added, and then the tube was shaken for 5 min. After centrifugation, each 500 μL of the aqueous and organic phases was subjected to γ -ray spectrometry with Ge detectors to determine the distribution ratios (D) of Fr and Cs. We also studied the extraction behavior of Fr and Cs with 1.5×10^{-3} M calix[4]arene-bis(benzocrown-6) in 1,2-dichloroethane from 10^{-4} – 3 M NaNO_3 / 10^{-4} M CsNO_3 aqueous solutions. We found that the D values of Fr and Cs change similarly as a function of the NaNO_3 concentration, though the D values of Cs are slightly higher than those of Fr in both the 10^{-4} – 3 M NaNO_3 and 10^{-4} – 3 M NaNO_3 / 10^{-4} M CsNO_3 solutions. In the conference, we will discuss these results by comparing with the literature data and those obtained with crown ethers.

P02 Production cross sections of ^{175}Hf in the $^{nat}\text{Lu}(p,xn)$ and $^{nat}\text{Lu}(d,xn)$ reactions

KOMORI, Y.¹, HABA, H.¹, AIKAWA, M.^{2,3}, SAITO, M.², TAKÁCS, S.⁴, DITRÓI, F.⁴

(¹RIKEN Nishina Center, ²Grad. School of Biomed. Sci. and Eng., Hokkaido Univ., ³Hokkaido Univ., ⁴ATOMKI)

A long-lived isotope of Hf, ^{175}Hf ($T_{1/2} = 70$ d), is useful for basic studies for rutherfordium (Rf, $Z = 104$). This isotope is producible in no-carrier-added form in the proton- and deuteron-induced reactions on ^{nat}Lu . However, excitation functions of these nuclear reactions have been scarcely studied. In this work, we measured the excitation functions of the $^{nat}\text{Lu}(p,xn)^{175}\text{Hf}$ and $^{nat}\text{Lu}(d,xn)^{175}\text{Hf}$ reactions up to 18-MeV proton and 24-MeV deuteron energies using a stack-foil technique and a γ -ray spectrometry. We performed these experiments at RIKEN and Institute for Nuclear Research (ATOMKI). The target stacks of Ta/Lu/Ti and Lu/Ti foils were irradiated for 2 h with proton or deuteron beams of approximately 180–240 nA. After the irradiation, each foil was subjected to γ -ray spectrometry with Ge detectors. We noticed that the half-life of ^{173}Hf is slightly longer than

that adopted in the current nuclear database. Therefore, we measured a precision half-life of ^{173}Hf in a separate experiment. In this work, we could measure the excitation functions of the $^{nat}\text{Lu}(p,xn)^{173,175}\text{Hf}$ and $^{nat}\text{Lu}(d,x)^{173,175}\text{Hf}$, $^{173,174m,174g,176m,177m,177g}\text{Lu}$ reactions. Thick-target yields of ^{175}Hf were also deduced from the measured excitation functions. The yields are 0.47 MBq/ $\mu\text{A}\cdot\text{h}$ at 17.2-MeV proton and 2.0 MBq/ $\mu\text{A}\cdot\text{h}$ at 24.0 MeV deuteron. We determined the half-life of ^{173}Hf to be 24.176 ± 0.012 h which is 0.58 ± 0.10 h longer than that in the database.

P03 Online anion-exchange experiment of ^{89m}Zr in HNO_3 for chemical study of Rf

WATANABE, E.¹, KASAMATSU, Y.¹, YOKOKITA, T.², HAYAMI, S.¹, TONAI, K.³, SHIGEKAWA, Y.², HABA, H.², SHINOHARA, A.¹

(¹Grad. Sch. Sci., Osaka Univ., ²Nishina Center, RIKEN, ³Sch. Sci., Osaka Univ.)

We are interested in the complexation of ^{104}Rf with nitrate ions and are planning to perform an anion exchange experiment of Rf using the extraction apparatus, AMBER. We have previously determined the experimental condition for Rf using long-lived radiotracers of homologue elements, ^{88}Zr , ^{175}Hf , and ^{234}Th . In this study, online anion-exchange experiments with the short-lived ^{89m}Zr ($T_{1/2} = 4.18$ min) in nitric acid were performed using a gas-jet transport system at the RIKEN AVF cyclotron.

^{89m}Zr was produced in the $^{89}\text{Y}(d,2n)^{89m}\text{Zr}$ reaction. The products were transported to AMBER with the He/KCl gas-jet system. We performed anion-exchange experiments of ^{89m}Zr using Adogen 464 resin in 4.3 – 8.1 M HNO_3 . The radioactivity of ^{89m}Zr was determined by gamma-ray spectroscopy with a Ge detector.

Although a part of ^{89m}Zr was adsorbed on the reaction container of AMBER, the K_d values, which were obtained by correcting the adsorption of Zr, are consistent with those in the batch experiment. In addition, we repeated the anion-exchange experiment 30 times by

automatic control, and constant K_d values were obtained throughout the experiment. From this study, it was confirmed that AMBER can be applied to the anion-exchange experiment of Rf in nitric acid.

P04 Coprecipitation experiment of ^{99}Mo with samarium hydroxide for chemical experiment of Sg

KASAMATSU, Y.¹, TONAI, K.¹, HAYAMI, S.¹, WATANABE, E.¹, TAKAMIYA, K.², SHINOHARA, A.¹

(¹Grad. Sch. Osaka Univ., ²KURNS)

In our previous study, we developed the coprecipitation method with samarium hydroxide to investigate the chemical behavior for superheavy elements. Indeed, we succeeded in investigating the coprecipitation behavior for element 104, Rf. The purpose of this study is to establish the coprecipitation experimental method including development of a new apparatus for the coprecipitation experiment of element 106, Sg.

We used carrier-free ^{99}Mo sample produced from fission of ^{235}U and macro amount of Mo sample including ^{99}Mo produced by neutron activation of Mo at KURNS. Coprecipitation behavior of Mo with samarium hydroxide and precipitation behavior of Mo were both investigated by conventional suction filtration method. In addition, Coprecipitation behavior of ^{99}Mo was also investigated using a fast and flow filtration equipment developed for online experiment of Sg. Consequently, the coprecipitation yield (~0%) which is consistent with precipitation yield under each condition was obtained for Mo, suggesting that the hydroxide precipitation properties of group 6 elements can be investigated through their coprecipitation behavior with Sm. The coprecipitation yields obtained with the apparatus were equivalent with those obtained by conventional filtration method. It was found that the developed equipment is applicable for Sg experiment.

P05 Development of sample data library for radioactive particles

SATOU, Y.¹, IGARASHI, Y.², MARTIN, P.G.³, ALHADDAD, O.³, SCOTT, P.G.³

(¹CLADS JAEA., ²KURNS Kyoto Univ., ³Univ. Bristol)

Radioactive particles so called Cesium micro particles is one of nuclear forensic material emitted from Fukushima Daiichi nuclear power plant accident, number of researchers in the world approaching these analyses for disclose particle generation process. However, key researchers of these particle in Japan will be retired, and these samples will be lost their whereabouts. We proposed sample archive system to protect the particles for the future. The system combined physical sample storage and these data library. The library works collecting basic sample information (when, where, who collected), ownership, lending history, also laboratory and system common sample IDs. The library store non-destructive and destructive analyses results such as α, β, γ -ray emitting radionuclides, SEM, EDS, synchrotron X-ray analyses results (XRT, XRD, XRF, XAFS), SIMS, ICP-MS and so on. Currently, the library works completely independent from each other's country due to export control and sample ownership in unpublished data, but finally the library will be open to public.

P06 The evaluation of ^{129}I internal exposure of infants due to intake of breast milk

OTA, T.¹, MATSUZAKI, H.², KODAMA, H.³, TERADA, H.⁴, NOMURA, K.⁵, OTA, Y.¹, WANG, X.¹, IIDA, M.¹, HIBINO, Y.¹, OKAYAMA, K.⁶, TOYA, M.², WATANABE, M.¹

(¹Japan chemical analysis center, ²The University of Tokyo, ³Teikyo Heisei University, ⁴National Institute of Public Health, ⁵Akita University, ⁶Hiroshima International University)
I-129 is a long-lived (Half-life: 15.7 million year) iodine radioisotope which is mainly produced by nuclear fission of Uranium. It has been discharged to environment from nuclear facilities

including fuel reprocessing plants and nuclear power stations. I-129 is concentrated in thyroid when taken into human body, causing long-term internal exposure which leads to a potential cause of thyroid cancer.

The purpose of this study is to assess the infant's internal exposure to I-129. For this purpose, we analyzed I-129 concentration in human milk which is a main source of nutrition for infants. To measure extremely low concentrations of I-129, AMS (Accelerator Mass Spectrometer) was adopted. In this study, we applied combustion methods to retrieve iodine quantitatively from human milk samples to overcome interference caused by high fat content. We established a new method to retrieve radioactive iodine in human milk samples quantitatively through combustion. Analysis of iodine 129 in the collected human milk was carried out, and the internal exposure to iodine 129 due to human milk intake was evaluated.

P07 Trends in Proficiency Tests Provided by IAEA ALMERA

OTA, T.¹, OTA, Y.¹, SATO, S.¹, SUZUKI, K.¹, NISHIMORI, C.¹, WATANABE, M.¹, ISOGAI, K.¹

(¹Japan Chemical Analysis Center)

The ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity) was established by IAEA in 1995 to share technologies for reliable and rapid analysis in emergencies. ALMERA consists of 187 laboratories representing 89 countries as of May 2020. We have been participating in the network since 2008. ALMERA's main activities are an implementation of proficiency tests, development of analytical methods, development of reference samples for radioactivity analysis, and implementation of workshops and training. Annual meetings are held once a year to actively exchange information. We use the ALMERA proficiency test as a part of our external quality control program. The proficiency test is held every year, and the contents of the program are updated every time. In this presentation, we will

present the results of recent IAEA ALMERA proficiency tests.

P08 Consideration on the Concentration Ratio of Strontium from Ambient Water to Aquatic Biota

TAGAMI, K., UCHIDA, S.

(Natl. Inst. Quantum Radiol. Sci. Technol.)

The amount of released ⁹⁰Sr was much smaller than those of radiocaesium from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident; yet, it is still necessary to assess the dose from the radionuclide because the radionuclide has long biological half-life in human body. Concentration ratio (CR) can be used to estimate radionuclide concentrations in foods; unfortunately however, internationally available CR data of Sr for aquatic biota were usually categorized in big groups, such as fish, molluscus, and algae. More detailed CR data for each species are helpful for reliable dose assessment. In this study, therefore, we carried out a data survey to enhance the Sr CR data sets for aquatic biota in marine and freshwater systems. In total, ca. 3700 CR data for aquatic biota were collated with species names and analyzed tissues (muscles, bones, whole etc.). For freshwater fish collated mainly in Japan, no time dependence of CR in common carp were found but for crucian carp, some increments were observed for the data obtained after the FDNPP accident. We found that the difference was mainly due to the co-present major elements in water which affect the uptake of Sr by fish body as previously reported outside Japan.

P09 Evaluation of uncertainty in gamma-ray spectrometry due to non-uniform distribution of radionuclides in sample container

SUZUKI, K., MIYATA, S., OHTA, Y., ISOGAI, K.

(Japan Chemical Analysis Center)

The measurement uncertainty in gamma-ray spectrometry with Ge semiconductor detector was evaluated due to non-uniform distribution of radionuclides in measurement sample.

Calculation of uncertainty was performed based on the framework of JCGM 101:2008 (ISO/IEC Guide 98-3/Supplement 1:2008) by using EGS5 Monte Carlo simulation code.

Firstly, numerical models were constructed for a coaxial p-type Ge semiconductor detector and a rectangular parallelepiped sample container with a cross section of $4.2\text{cm} \times 4.2\text{cm}$ square. Secondly, the sample in the container was virtually divided into small cubes with a side length of 2 mm, then the 10% of the cubes were randomly selected as “gamma-ray sources” and the counting efficiencies (count/number of photons) for the collection of the selected sources were calculated. Finally, the uncertainty was evaluated from the frequency distribution of counting efficiencies obtained by repeating this process. The conditions for the simulation were as follows: (sample filling height: 10,20,30,40,50mm, gamma-ray energy: 88.8keV, 661.7keV, 1836.2keV, number of repetitions: 10,000)

The uncertainty due to the distribution of radionuclides in the measurement container was estimated within 2.5%.

P10 Isotopic ratios of plutonium in the radioactive particles released by the accident of Fukushima Daiichi Nuclear Power Plant.

IGARASHI, J.¹, ZHENG, J.², ZHANG, Z.¹, NINOMIYA, K.¹, SATOU, Y.³, FUKUDA, M.², NI, Y.^{2,4}, AONO, T.², SUEKI, K.⁵, SHINOHARA, A.¹

(¹Graduate School of Science, Osaka University, ²QST, ³JAEA, ⁴School of Physics, Peking University, ⁵Graduate School of Pure and Applied Sciences, Tsukuba University.)

Plutonium is one of the most notable radionuclides in the nuclear accident. The amounts of Pu released by the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident were investigated so far. However, because the released amounts of Pu isotopes is small compare to global fallout, it is difficult to accurately evaluate Pu contamination caused by the FDNPP accident. On the other hand, Pu contamination

source can be investigated from Pu isotopic ratios since Pu isotopic ratios are different by Pu contamination sources of FDNPP accident, Chernobyl accident and global fallout. In this work, we focused on the insoluble particles having highly concentrated radioactive Cs (radioactive particles) released from the FDNPP to determine accurate Pu isotopic ratios. Due to their insolubility, the particles retained the original property at the time of emission from the reactor, and have never contaminated by Pu from global fallout. We identified Pu isotopes for 6 radioactive particles by radiochemical analysis and ICP-MS measurement. The average values of $^{240}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratios in the radioactive particles were determined as 0.383 ± 0.019 and 0.171 ± 0.012 , respectively. These values were slightly different from the calculated value of the core inventory.

P11 Study on prevention effect of radionuclides adsorbed on the surface of measurement containers

KONNO, Y.¹, MAEYAMA, T.¹, SAEGUSA, J.², SHINOHARA, H.¹, OTA, H.¹, ISOGAI, K.¹
(¹Japan Chemical Analysis Center, ²Japan Atomic Energy Agency)

It is well known that radioactive cesium and radioactive iodine are adsorbed on the measurement container's surface. In this study, we used some reagents in measurement vessel trying to find an anti-adsorption method. The study was conducted at the time of revising “The series of environmental radioactivity measuring methods No.24 (Sample Pretreatment for Gamma-ray Spectrometry in Emergencies)”, to confirm the prevention effect of radionuclide-adsorption radionuclides adsorbed on the surface. In our study, we prepare four 2L Marinelli beakers, and pour 2L tap water. Three beakers were added to sodium chloride, sodium thiosulfate and nitric acid, respectively. The other one was nothing added as a comparative control. After these preparatory works, we added I-131 and Cs-137 standard solutions in these beakers. We measured these units by germanium

semiconductors detector immediately after the standard solution added. The concentrations of the solutions were repeatedly measured to observe their chronological changes. As a result, the adsorption of Cs-137 to the surface was not found in any units, whereas that of, the adsorption of I-131 was found at each of three beakers with sodium chloride, nitric acid, and the control. We did not find the adsorption of I-131 to surface in the sodium thiosulfate unit. Based on the above, the sodium thiosulfate is designated as an additive for measuring radioactive iodine in the revised “The series of environmental radioactivity measuring methods No.24”

P12 Quality assurance in environmental radioactivity monitoring.

ABE, G., OHTA, H., KAWAMURA, T., KONNO, Y., TOYOOKA, S., SUZUKI, K., SATO, S., OTA, T.

(Japan Chemical Analysis Center)

After the accident at TEPCO's Fukushima Daiichi Nuclear Power Plant, more severe attention is being paid to the analytical data on radioactivity, and it is required to provide more reliable data. There is environmental radioactivity monitoring by the Nuclear Regulation Department conducting nationwide. Japan Chemical Analysis Center (JCAC) is outsourced from the Nuclear Regulation Department, to ensure the reliability of radioactivity monitoring in Japan, we are conducting interlaboratory comparisons based on ISO/IEC 17043 for analysis institutions in all prefectures of Japan. Here, we report the interlaboratory comparison's results using by γ -ray spectrometry analysis conducted in FY2019.

JCAC prepares reference samples and distributes them to the analysis organization. The analysis organization analyzes and measures the reference samples. JCAC compares and evaluates the analysis and measurement results of analytical institutions with the assigned values. The evaluation method was calculated by the robust method using Z-score.

According to evaluation results, a higher accurate analysis was performed in the interlaboratory comparisons. The causes of the unsatisfactory results were investigated and through reanalysis and reanalysis, and the improvements were also observed.

P13 Ecological cesium resuspension generated by precipitation-Coarse bioaerosol

IGARASHI, Y.¹, KINASE, T.², KITA, K.³, HAYASHI, N.³, ISHIZUKA, M.⁴, ADACHI, K.², SEKIYAMA, T.-T.², KOITABASHI, M.⁵, ONDA, Y.⁶

(¹Kyoto Univ., ²Meteorolo. Res. Inst., ³Ibaraki Univ., ⁴Kagawa Univ., ⁵Nat. Inst. Agro-Environ. Sci. & ⁶Univ. Tsukuba)

Even though it has been nine years since the Fukushima Dai-ichi Nuclear Power Plant (F1NPP) accident, detectable levels of F1NPP radiocaesium (10^{-5} to 10^{-3} Bq m⁻³) are still present in the air over evacuating zone of Fukushima Pref., where was the most contaminated region. The continued detection of radiocaesium is attributable to its resuspension, but the mechanism of that resuspension is not well understood. In the present study, we add additional evidence for this rain-enhanced/induced aerosol emission in a forest environment, that is outbreak of radiocesium-bearing aerosols in a Japanese forest due to precipitation during warm season. We confirmed that the fungal spore sources in the rainy weather for the radiocesium resuspension seem to be different from those in the fine weather, by applying the weather dependent sample collection. Larger fungal particles (possibly conidia and ascospore) are probably emitted during precipitation than during fine weather, suggesting that the rain-splash mechanism for mold-like fungi could be the major mechanism.

P14 Simulated generation experiment by laser irradiation for radioactive particles emitted from the Fukushima nuclear accident

SATO, R.¹, INAGAKI, M.², SEKIMOTO, S.², TAKAMIYA, K.², OKI, Y.², OHTSUKI, T.²
(¹Kyoto Univ., ²KURNS)

A large amount of radioactive materials was released into the environment as a result of the Fukushima nuclear accident. There are variety of forms of the radioactive materials. For instance, gas, aerosol, and fine particles. Because the physical and chemical forms of the released radioactive materials affect the deposition and migration in the reactor and in the environment, the elucidation of their dynamics requires information on the initial state of them at the time of formation and release. Therefore, we decided to simulatly generate the radioactive particles by laser irradiation. Siliceous material was processed by laser irradiation for generating simulated particles. Irradiation produced the fume and it was captured by impactor. As a result of SEM observation of the fume, a several particles were discovered. The sizes of particles weve 1 to 20 μm and the shape was spherical. Accordingly, the experimental apparatus used in this study is able to generate simulated particles sufficiently. The examination for the simulated generation experiment was also undertaken with other nominated materials and compared with the actual particles. The details result of them will be reported in the presentation.

P15 Distribution of ⁹⁰Sr, ¹³⁷Cs, and direction dependency of ⁹⁰Sr/¹³⁷Cs around the FDNPP

KAJIWARA, T.¹, NINOMIYA, K.¹, SHINOHARA, A.¹, YAMAGUCHI, Y.²

(¹Osaka Univ., ²Institute for Radiation Sciences, Osaka Univ.)

In the nuclear accident, ¹³⁷Cs and ⁹⁰Sr are the most important nuclides because of their large core inventories and long half-lives. However, the researches on environmental behavior of ⁹⁰Sr released by the accident of FDNPP are limited compare to these of ¹³⁷Cs due to the difficulty on quantification. Release process of ⁹⁰Sr and ¹³⁷Cs were strongly influenced by their volatility properties; ⁹⁰Sr is low-volatile element, and ¹³⁷Cs is high-volatile element, as a result, ⁹⁰Sr/¹³⁷Cs

was changed by core temperatures. In previous studies, when the core temperature reached 2000K, ⁹⁰Sr/¹³⁷Cs becomes 10^{-4} . From deposited ⁹⁰Sr/¹³⁷Cs distribution, we can categorize release events from the FDNPP and investigate release process and core temperature. In this work, we analyzed ⁹⁰Sr and ¹³⁷Cs in 21 soil samples collected around FDNPP. After extraction of ⁹⁰Sr from the soil sample by solid phase extraction technique ⁹⁰Sr activity was determined by liquid scintillation counter. Chemical yield was identified by ICP-MS measurement. Concentration of ¹³⁷Cs was 4.8-294 Bq/g, and that of ⁹⁰Sr was 0.41-34 mBq/g. ⁹⁰Sr/¹³⁷Cs changed by direction from FDNPP. In north to northwest and south direction, ⁹⁰Sr/¹³⁷Cs was order of 10^{-4} , it indicated core temperature reaches around 2000K. In west direction, ⁹⁰Sr/¹³⁷Cs was order of 10^{-5} .

P16 Effect of chemical characteristics of fission products on generation process of radioactive aerosols

TAKEUCHI, T.¹, TAKEUCHI, Y.¹, TAKAMIYA, K.², INAGAKI, M.², SEKIMOTO, S.², OKI, Y.², OHTSUKI, T.²

(¹Graduate School of Engineering, Kyoto Univ.,

²Institute for Integrated Radiation and Nuclear Science, Kyoto Univ., Osaka Japan)

The generation mechanism of the radioactive aerosol has been studied by developing an apparatus that simulates the generation process of radioactive aerosol containing fission products (FP) in our previous work. The apparatus consists of four parts; generation of aerosol, supply of FPs, mixing chamber and collecting filter. In order to supply FPs, neutron irradiated uranium sample had been used. In the present study, the irradiation condition was changed to observe the generation mechanism of different FPs because chemical effect of FP in the generation process of radioactive aerosols might become clear. As a result, it was found that FPs of ⁸⁷Kr, ⁹²Sr, ⁹⁴Y, ¹⁰⁵Ru, ¹²⁸Sb, ¹³¹Te, ¹³³mTe, ¹³⁴Te, ¹³²I, ¹³⁴I, ¹³⁵Xe, ¹³⁸Cs, ¹⁴²La are available to observe their behaviors by shortening irradiation and standing

times. Plural isotopes were observed for ruthenium, tellurium and iodine that enables to check the reproductivity of experiments by comparing the behavior of FPs among isotopes. The attachment behavior of FPs to solution aerosols will be discussed by observed attachment ratio of these FPs.

P17 Strontium-90 measurement in trace amount of hard tissues for distribution analysis

KOARAI, K.¹, MATSUEDA, M.^{1,2}, AOKI, J.^{1,2}, YANAGISAWA, K.², FUJIWARA, K.¹, TERASHIMA, M.¹, KINO, Y.³, OKA, T.¹, OKUTSU, K.³, YAMASHITA, T.³, TAKAHASHI, A.³, SUZUKI, T.³, SHIMIZU, Y.³, CHIBA, M.³, OSAKA, K.³, SASAKI, K.³, SEKINE, T.³, FUKUMOTO, M.^{3,4}, SHINODA, H.³, KITAMURA, A.¹, ABE, H.¹

(¹JAEA, ²Fukushima Univ., ³Tohoku Univ., ⁴Tokyo Med. Univ.)

We demonstrated a method of ⁹⁰Sr measurement in small pieces of hard tissues with radioactivity measurement or ICP-MS measurement. Interference elements of the measurements were removed by chemical separation. We could determine ⁹⁰Sr in 0.1 g of hard tissues by radioactivity measurement method and ICP-MS method. Limit of detection of the ICP-MS method was lower than that of the radioactivity measurement. The ICP-MS method is adequate method for distribution analysis of ⁹⁰Sr in the hard tissues.

P18 Sample preparation procedure for ESR dosimetry using teeth of wild animal

OKA, T.¹, MITSUYASU, Y., TAKAHASHI, A., KOARAI, K., KINO, Y., SEKINE, T., OKUTSU, K., YAMASHITA, T., SHIMIZU, Y., CHIBA, M., SUZUKI, T., OSAKA, K., SASAKI, K., FUJISHIMA, Y., URUSHIHARA, Y., GOH, V., ARIYOSHI, K., NAKATA, A., YAMASHIRO, H., SUZUKI, M., FUKUMOTO, M., MIURA, T., SHINODA, H.

(¹JAEA, ²Tohoku Univ., ³Hirosaki Univ., ⁴Fukushima Medical Univ., ⁵Hokkaido Univ. of Science, ⁶Niigaka Univ, ⁷RIKEN)

Releases of the radioactive materials from the Fukushima Daiichi Nuclear Power Plant accident result in a low dose exposure to wild animals. The external dose of wild animals is commonly estimated by the external dose rate of the captured point, but the estimated exposure dose has huge uncertainties because the size of the habitat and/or the movement of the wild animals are not included in the estimation. To estimate the external dose precisely, we utilize electron spin resonance (ESR) dosimetry. In this work, we investigated the sample preparation procedure of wild animals. The ESR spectrum of enamel of racoon captured in Namie-town, Fukushima has a broad ESR signal due to metal components. We attempted to remove metal components by chemical treatments. After the chemical treatment, the irradiation of ⁶⁰Co gamma-ray and ESR measurements were repeated to obtain the relationship between the CO₂ radical intensity and the absorbed dose. The linear relationship between them suggests that we can apply ESR dosimetry for racoon teeth. Using this relationship, the external exposure dose was estimated to be 68.8 mGy.

P19 Surface water circulations in the southwestern Sea of Japan: Implications from the Ra-228/Ra-226 ratio and Cs-134 concentration

HANAKI, S.¹, INOUE, M.¹, TAKEHARA, R.¹, MOROKADO, T.¹, SHIROTANI, Y.¹, TAKIKAWA, T.², YOSHIDA, M.³, OKINO, A.⁴, MIKI, S.⁵, NAGAO, S.¹

(¹Kanazawa Univ., ²Nagasaki Univ., ³Shimane Univ. Oki Marine Biological Station, ⁴Shimane Prefecture, ⁵Fisheries Resources Institute)

The concentration (activity) ratios of radium isotopes (²²⁸Ra/²²⁶Ra ratio) at the surface of the southwestern Sea of Japan showed high values (1.5–2.5) in July–December (2015–2019), indicating a seasonal variation. This was due to the high mixing ratio of ²²⁸Ra-rich continental

shelf water from the western East China Sea (ECS). Furthermore, the $^{228}\text{Ra}/^{226}\text{Ra}$ ratios of the waters exhibited a wide lateral variation with higher values relative to those in coastal areas (1.5–2.0). The total fractions of waters from the ECS continental shelf and Yangtze River (f_{S+Y}) in the southwestern Sea of Japan (0.2–0.5), based on ^{228}Ra concentration and salinity, were 1–1.5 times higher than those of the coastal waters (0.2–0.3), in contrast to a small variation with lower f_{S+Y} values in January–June (particularly in March–June). This indicated the irregular patterns of the lateral intrusion of the ^{228}Ra -rich branch of the Tsushima Warm Current in July–December, passing through of the Tsushima Strait.

P20 Method development for simultaneous determination of ^{237}Np and Pu isotopes in seawater by SF-ICP-MS with a single column chromatographic separation

ZHANG, S.^{1,2}, YANG, G.¹, ZHENG, J.¹, PAN, S.², AONO, T.¹

(¹ National Institutes for Quantum and Radiological Science and Technology, ² Nanjing Univ.)

Plutonium in seawater is a well-known tracer in biogeochemical processes with its particle-reactive properties. Recent studies evidenced that another transuranic isotope, ^{237}Np ($t_{1/2}=2.14\times 10^6\text{y}$) has a similar behavior of ^{137}Cs , thus shows great potential to act as a tracer for water mass circulation for decadal and longer time scales. However, information on ^{237}Np presented in seawater, especially in open ocean is very scarce, the distribution and inventory of ^{237}Np in the ocean remains largely unknown due to its low concentration in seawater and great analytical challenge for its measurement. Most methods for analysis of ^{237}Np need a large volume seawater, tedious chemical separations, and long counting times in the radiometric methods of alpha spectrometry, and the expensive AMS devices. In this study, we present a new method for simultaneous determination of ^{237}Np and Pu isotopes in seawater by SF-ICP-MS.

^{242}Pu was employed as a non-isotopes yield tracer for ^{237}Np and Pu isotopes. A single column anion-exchange (AG MP-1M) chromatography was used for chemical separation of ^{237}Np and Pu isotopes from seawater matrix. This method was applied to determine the seawater samples collected from the Pacific Ocean for the first time.

P21 Re-suspension of insoluble Cs radioactive particles to the atmosphere

TANG, P.¹, HATANAKA, K.¹, KITA, K.¹, SATOU, Y.², NINOMIYA, K.³, SHINOHARA, A.³, KINASE, T.⁴, ADACHI, K.⁴, IGARASHI, Y.⁵

(¹Ibaraki Univ., ²CLADS/JAEA, ³Osaka Univ., ⁴MRI, ⁵Kyoto Univ.)

Due to the Fukushima Daiichi Nuclear Power Plant accident (FDNPP), insoluble radioactive Cs particles were generated and released into the environment. Resuspension of such particles, which may be derived from FDNPP decommissioning work, are recognized as a long-term potential risk factor. In this study, we investigated the reemission-possibility and the physicochemical characteristics of these insoluble radioactive Cs particles. Twenty radioactive Cs particles were detected in 186 aerosol samples collected from 2015 to 2019 in a difficult-to-return-zone about 25 km far from FDNPP. SEM-EDS analysis was performed on the successfully isolated 15 Cs bearing particles, and all were type-A particles with the diameters of 1–2 μm . Although the resuspension-frequency was low, it can be clarified that such radioactive Cs particles could be re-emitted to the atmosphere.

P22 Speciation of uranium particles using $\mu\text{-X-ray}$ absorption near edge structure spectra

YOMOGIDA, T.^{1,2}, OUCHI, K.², KITATSUJI, Y.², TAKAHASHI, Y.¹

(¹Tokyo Univ., ²JAEA)

Micro X-ray absorption near edge structure (XANES) spectroscopy was applied to the

characterization of uranium particles with a diameter of a few micro-meter. Uranium dioxide(UO_2), Uranium peroxide($\text{UO}_4 \cdot n\text{H}_2\text{O}$), Uranyl nitrate ($\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) were employed. Uranium samples were diluted in boron nitride for XANES measurement performed at BL01B1 of SPring-8. Uranium particles on carbon tape were used for micro-XANES measurement at BL37XU of SPring-8.

The XANES spectrum of the UO_2 particle with a diameter of 3 μm was consistent with the standard UO_2 spectra. The result is showing the applicability of the proposed methods to analyzing micron sized uranium particle. However, a large spectral distortion was observed in the XANES spectrum of the small uranium particles with a diameter less than 3 μm . X-ray beam shift in the measurement or background from Br in the carbon tape would be interfering with micro-XANES measurement. Improvements of sample preparation and micro-XANES measurement method is indispensable for speciation of small uranium particles.

P23 Feasibility study on the on-line monitoring of tiny particles containing alpha-emitters generated in the fuel-debris retrieval from the reactors of Fukushima Daiichi Nuclear Power Plant

TOYOSHIMA, A., FURUTANI, H., TERAMOTO, T., MUTA, H., TOKOI, K., YOSHIMURA, T., TOYODA, M., SHINOHARA, A.

(Osaka Univ.)

The retrieval of the fuel debris from the reactors in Fukushima Daiichi Nuclear Power Plant is planned to start at FY2021. It is concerned that radioactive tiny particles containing alpha-emitters such as ^{238}U and ^{239}Pu are generated in the cutting process of the debris and are dispersed to outside of the reactors. We, therefore, suggest on-line monitoring of the tiny particles using an Aerosol Time-Of-Flight Mass-Spectrometer (ATOFMS) to prevent the

particles from leaking out of the reactors in the debris retrieval. At present, we are preparing the generation apparatuses of tiny particles from modeled solid and liquid samples and the on-line detection system using ATOFMS. In this study, we present the present status of the sample preparation, developments of the generation apparatuses, and results of preparatory experiments.

P24 Non-destructive image experiment using Muonic X-ray with Si Double-sided Strip Detector

CHIU, I-H.¹, KAJINO, M.¹, SHINOHARA, A.¹, NINOMIYA, K.¹, TAKEDA, S.², KATSURAGAWA, M.², TOMARU, R.², YABU, G.², NAGASAWA, S.², TAKAHASHI, T.², WATANABE, S.³

(Osaka University¹, Kavli IPMU², JAXA³)

This study presents an element analysis method using a muon beam. Muon stopped in the matter and they are captured by element. When muon transitions to a low energy level in atom, muons characteristic X-rays are radiated. This makes it possible to non-destructively analyze the elements of matter with an excellent energy resolution detector. Furthermore, we will develop a two-dimensional silicon detector with high position resolution. It is possible to reconstruct the image by muon characteristic X-rays with this two-dimensional detector. When the muon characteristic X-rays come into a silicon semiconductor, lots of electron and hole are produced. The amount of electric charge is calculated for the energy of the input muon characteristic X-rays. Then the energy spectrum with high resolution can be obtained. At the same time, the double-sided strip detector provides position information of detector with high position resolution. Combining the energy spectrum and position information, an image of the matter is reconstructed. This talk will introduce the development and details of the silicon double-sided strip detector. Then, the developed silicon detector was subjected to a muon X-ray experiment at the J-PARC.

P25 Mössbauer Characterization of Iron Oxidation State during Carbonization of Iron-Loaded Woody Biomass

YAMAUCHI, S.¹, YAMAGISHI, T.², KURIMOTO, Y.¹, SUZUKI, H.¹, TAKAYAMA, T.³, SAKAI, Y.³

(¹Akita Prefectural Univ., ²Seikei Univ., ³Daido Univ.)

Recently, woody biomass has attracted attention as the resources of functional carbon materials. Conductive filler made from iron-loaded charcoal is expected to be put into practical use in terms of high electric conductivity and low cost. The iron-loaded woody charcoal is graphitized from considerably low temperatures; however, the whole mechanisms of this carbonization reaction has not been completely explained. In particular, many of details of iron-role in carbonization are not elucidated. We investigated the effects of carbonization temperature and cooling rate on the chemical states of iron and carbon using Mössbauer and Raman spectroscopy in order to discuss the mechanism of the changes in iron-oxidation states.

P26 Hyperfine field measurements for the investigation of nuclear spin relaxation of the A site probes in CdFe₂O₄ by means of ⁵⁷Fe Mössbauer spectroscopy

ITO, T.¹, KONAKA, M.², SATO, W.^{1,2}

(¹Grad. Sch. Nat. Sci. Tech., Kanazawa Univ., ²Inst. Sci. Eng., Kanazawa Univ.)

Spinel oxides exhibit various physical properties depending on the constituent metal elements and on their residence positions: the tetrahedral A site and octahedral B site. Especially, electric conductivity and magnetic property of spinel oxides have been extensively studied for practical application. In our recent study, hyperfine fields in CdFe₂O₄ were measured by means of perturbed angular correlation spectroscopy with the ¹¹¹Cd(←^{111m}Cd) probes and obtained spectra implying nuclear spin relaxation of the probe occupying the A site at two different temperatures (room temperature and 77 K).

However, the cause of this phenomenon is still unknown. In order to provide insight into the unexpected phenomenon, we measured hyperfine fields in Cd_xFe_{3-x}O₄ (x=0.25, 0.5, 0.75, 1.0) by means of ⁵⁷Fe Mössbauer spectroscopy to investigate the local environment of Fe nuclei. Magnetic interactions appearing in the spectra gradually disappear as the Cd concentration increased, and the fitting results revealed that Cd occupies the A site for all the samples investigated. The results are consistent with those of the previous studies, suggesting that the nuclear spin relaxation phenomenon is a characteristic of Cd at the A site.

P27 ⁵⁷Fe Mössbauer spectra of sand samples of Inamuragasaki in Kamakura

YOSHIDA, M.¹, ENDO, K.², KOBAYASHI, Y.¹
(¹University of Electro-Communications, ²Showa Pharmaceutical University)

Iron is contained in iron oxides, iron sulfides, and the substituted positions of metal-ions in silicates in the crust. It is important to study the chemical forms of Fe atoms in environmental samples from the view point of earth science and environmental science. There are many beaches in Shonan area near Kamakura. Only the Inamuragasaki beach has a color of black. ⁵⁷Fe Mössbauer spectroscopy was applied to the sand of Inamuragasaki beach in order to characterize the chemical form of Fe atoms. ⁵⁷Fe Mössbauer spectra of the sand samples were measured at RT, 77 K and 15 K, respectively. X-ray diffraction patterns of them were also observed.

The Mössbauer spectra of sand samples obtained at RT could be analyzed by two components of a doublet and a singlet. One of the results was demonstrated to be a doublet with *I.S.* (δ) = 1.31(1) mm/s and *Q.S.* = 2.35(2) mm/s and a singlet (S) with δ = 0.29(4) mm/s. It is considered that a doublet and a singlet is assigned to be in Fe²⁺ (H.S.) and Fe³⁺ (H.S.), respectively. Since there is clearly no magnetic splitting, it was determined to be in paramagnetic states. Based on ⁵⁷Fe Mössbauer spectroscopy and XRD, the

chemical forms of Fe atoms in sand samples of Inamuragasaki will be discussed.

P28 Antimony-121 Mössbauer spectra of square pyramidal organoantimony compounds with catechol ligands

TAKAHASHI, M., IITSUKA, T.

(Toho Univ.)

Mössbauer spectra for the square pyramidal (SP) organoantimony R_3SbL ($R = Ph, p\text{-FC}_6\text{H}_5, p\text{-Tol}, o\text{-Tol}, o\text{-Xyl}, Mes$) with catechol (H_2cat) and 2,3,4,5-tetrachlorocatechol ($H_2Cl_4\text{-cat}$) were measured at 20 K and the electron distribution was compared with those of the TBP complexes. The isomer shift δ (relative to InSb) was 3.66 to 4.32, the quadrupole coupling constant e^2qQ was -8.56 to -12.2 $mm\ s^{-1}$, and the asymmetry parameter η 0.24 to 0.66. The value of e^2qQ was smaller than the value of -20.6 $mm\ s^{-1}$ of $Ph_3Sb(OTf)_2$ with TBP structure, which was about half. This strongly reflects the difference in structure. We calculated the Muettertues and Guggenberger's dihedral diagonal δ_{24} , which evaluates the SP character of the five-coordination complex, to examine the relation with the Mössbauer parameters. The e^2qQ tends to decrease according to δ_{24} . Consequently, the SP structure was found to give different parameters than TBP, and the electron distribution was, however, also strongly influenced by the substituents.

P29 VRFB membrane permeability tests using radiotracer ^{48}V

SHIRASAKI, K.

(IMR, Tohoku Univ.)

Vanadium redox-flow battery (VRFB) draws attention for the energy storage system, which is particularly useful for conjunction with renewable energy sources. The VRFB consists of electrodes, membranes, electrolytes and only vanadium ions as active substance in both positive (VO^{2+}/VO_2^+) and negative (V^{3+}/V^{2+}) electrolytes [1]. This constitution makes it convenient to recover battery capacity to initial state by adjusting the concentration and valence

of vanadium ions between positive and negative electrolytes, and makes it difficult to directly evaluate vanadium ions crossover of the membrane, simultaneously.

In the last year, we developed an evaluation method of the crossover of vanadium ions through the membrane using radiotracer ^{48}V [2]. The method has a potential for understanding nature of the membrane both in a self-discharge and coulombic efficiency during charge-discharge processes of the VRFB.

In this study, we investigate the behavior of crossover of vanadium ions in the different types of the membranes.

[1] E. Sum, M. Skyllas-Kazacos, J. Power Sources, 15 (1985) 179-190.

[2] K. Shirasaki, T. Yamamura, J. Membr. Sci., 592 (2019) 117367.

P30 Development of ^{211}At labeling method by electrolysis.

NAKAGAWA, S., KADONAGA, Y., TOYOSHIMA, A., OOE, K., KANDA, A., NAGATA, K., TERAMOTO, T., TOKOI, K., YOSHIMURA, T., SHINOHARA, A.

(Osaka Univ.)

^{211}At (half-life = 7.2 h) is expected to be used for targeted α -therapy. There are various methods for labeling ^{211}At on molecular targeting agents. The substitution reaction of ^{211}At using a harmful metal such as tin or mercury is common. However, it is concerned to be problematic for human health in clinical uses. Therefore, in this study, we aim at labeling ^{211}At by a clean method by applying electrolysis that does not contain any oxidants and metals. As a labelled compound, astato- α -methyl-L-tyrosine ($^{211}At\text{-AAMT}$), which is currently labeled by substitution using mercury, was selected. Because it is important to generate ^{211}At cations for the electrolytic labeling reaction, we first aimed to change the chemical species of astatine. In the experiment, the electrolytic oxidation of ^{211}At was carried out under various conditions of pH and applied potentials. Formed ^{211}At species were assayed by Thin Layer Chromatography (TLC) before and

after the electrolysis. Spot positions of ^{211}At were observed using Imaging Plate. After confirming change of oxidation states of ^{211}At before and after the electrolysis, an electrolytic labeling reaction was performed. As a result, oxidation states of astatine species were changed after electrolysis. The electrolytic labeling reaction will be reported in the symposium.

P31 Development of identification method for chemical form of astatine using mass spectrometry

TOKOI, K., TOYOSHIMA, A., KADONAGA, Y., TERAMOTO, T., SHINOHARA, A.

(Osaka Univ.)

^{211}At is considered as one of the most promising radionuclides for targeted alpha therapy because of its suitable half-life of $t_{1/2} = 7.2$ h and α -particle emission probability of 100% including that of its short-lived daughter nucleus ^{211}Po . However, due to the lack of long-lived radioisotopes of At, its chemical properties have been still incompletely understood. In particular, for medical application, it is important to understand chemical forms of At under various conditions. Mass spectrometry is promising for the determination of chemical forms of At. In the At case, however, sample condensation is realistically important because of the limitation of its available amount. Therefore, we started at studying gold nanoparticles which are known to adsorb astatine quantitatively, as a condensation absorbent (matrix) for At. In this study, we report the development plan of the mass spectrometer for astatine and a sample concentration method for a lighter homolog of At, iodine. Details of results will be presented in the symposium.

P32 Production of ^{47}Sc for cancer therapy with accelerator neutrons by deuterons

HASHIMOTO, K.¹, TSUKADA, K.^{1,2}, SAEKI, H.^{1,3}, KAWABATA, M.^{1,3}, OHTA, A.^{1,3}

(National Institutes for Quantum and Radiological Science and Technology¹, Japan Atomic Energy Agency², Chiyoda Technol Corporation³)

Scandium-47 is an attractive radionuclide for cancer therapy, because of emissions of medium energy beta particle (a mean energy of 162 keV) and gamma rays (159 keV) suitable for imaging, and its appropriate half-life (3.35 days). In this study, we have investigated the production of ^{47}Sc by the $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ reaction using fast neutrons from $^{nat}\text{Be}(d,n)$ reaction at the CYRIC of Tohoku university. The irradiated natural Ti metal sheets were dissolved in HCl/HF (400/1) and ^{47}Sc as well as $^{44\text{m}}, 46, 48\text{Sc}$ and ^{47}Ca were detected in the solution. The purified ^{47}Sc was obtained by the DGA normal resin separation method with high recovery yield.

P33 Chemical separation of As-74 using solvent extraction and column chromatography

IKEDA, H.^{1,2}, KIKUNAGA, H.², KOHDA, T.³, QIAN, Z.J.³, CHIEN, M.F.³, INOUE, C.³, SUZUI, N.⁴, KAWACHI, N.⁴, RAMDHANI, D.¹, WATABE, H.¹

(¹CYRIC, Tohoku University, ²ELPH, Tohoku University, ³GSES, Tohoku University, ⁴QST)

We produced As-74 ($T_{1/2} = 17.77$ d), which is one of the positron-emitting nuclides of As, and investigated the chemical separation method. We produced As-74 by irradiating Ga_2O_3 pellet with 30 MeV alpha particles using the AVF cyclotron of Cyclotron and Radioisotope Center (CYRIC), Tohoku University. The irradiated Ga_2O_3 pellet was dissolved into 1 mL of 12 M NaOH solution while heating. After neutralization with about 2 mL of 6 M HCl, solvent extraction was performed with 5 mL of organic solvents containing bis(2-ethylhexyl)phosphate (HDEHP). The concentration of HDEHP was 1 M, and organic solvents were benzene, toluene, ethyl acetate, and hexane. From the ratios of As-74 in aqueous and organic phases, we selected hexane as the organic phase. Ethanol was added to the aqueous phase to 50 mL, and solid NaCl was precipitated. The supernatant was charged into an anion exchange column, and As-74 was eluted by 0.1 M HCl. The results of column chromatography suggested that almost all of

As-74 was eluted by 8 mL of 0.1 M HCl. From these results, we determined the chemical separation method of As-74 from irradiated Ga₂O₃.

P34 Development of a synthetic method for an aromatic hypervalent astatine compound

KANDA, A.¹, NAGATA, K.², TOYOSHIMA, A.³, SHINOHARA, A.^{1,3}, YOSHIMURA, T.²

(¹Grad. School of Sci., Osaka Univ., ²Radioisotope Res. Ctr., Inst. of Rad. Sci., Osaka Univ., ³Inst. of Rad. Sci. Osaka Univ.)

Astatine-211 is one of the promising nuclides for application to targeted alpha therapy. However, medical application of astatine-211 has been restricted by a lack of stability of astatine-labeled compounds. Astatine-211 is currently labeled to a molecule as an aromatic monovalent astatine, and trivalent (hypervalent) astatine is potentially suitable as an astatine-labeled compound because trivalent astatine ion can be incorporated in a cyclic planar three-coordinated structure. Surprisingly, chemical property of hypervalent astatine compound has not been studied. The study on influence electric and/or steric effects of a ligand to the hypervalent astatine compound is expected to lead to a novel stable astatine-labeled compound or elucidation of the chemical property of an aromatic astatine compound. In the present study, synthesis of (diacetoxyastato)benzene in which astatine is trivalent was attempted. Astatobenzene was oxidized with sodium hypochlorite in the presence of acetic acid. After the reaction, the astatine species in the reaction solution was analyzed with radio-HPLC. As the result, a heavy-tailed peak was observed at retention time (Rt) = 8.5 min which was different from 4.2 min of (diacetoxyiodo)benzene. The peak position changed to Rt = 12.5 min by addition of sodium sulfite to the solution after oxidation of astatine. The result suggests that the oxidized At compound was reduced by sodium sulfite to reproduce astatobenzene. It is now projected the experiments using radioiodine and astatine to change in electronic property of ligand to find out

the difference of Rt between astatine and iodine compounds.

P35 Synthesis of Radiometal Complexes Coordinated with New Macrocyclic Ligand Bearing Phosphonate Pyridine Arms

BABA, K.¹, NAGATA, K.², YAJIMA, T.³, YOSHIMURA, T.²

(¹Osaka Univ., ²Osaka Univ. RI center, ³Kansai Univ.)

In recent years, targeted alpha therapy (TAT) has attracted much interest. ²²⁵Ac is one of suitable nuclides for TAT. The reaction of the metal ion with a ligand is preferred to proceed in a mild condition to give a highly stable metal complex. In this study, we newly designed and synthesized the new ligands, macropp and macropp^{Et} in which two arms with phosphonic acid or phosphonic acid ester groups are attached to the 18-membered macrocycle, 4,13-diaza-18-crown-6. In addition, the lanthanum complexes using stable isotopes were synthesized. The *R_f* values of the ligands and the mixtures of La(III) and ligands were different one another in thin layer chromatography with adsorbent of alumina and cellulose. This indicates that the reactions of Ln(III) with one molar equivalent of the ligands gave the La(III) complexes in high yields at room temperature. The results of the reaction of Ac(III) with the ligands will be reported in the JNRS annual meeting.

P36 Production of Sc-46 and Sc-47 tracers by photonuclear reactions

KIKUNAGA, H.

(ELPH, Tohoku Univ.)

Scandium-46 and scandium-47 are nuclei that are useful or expected to be useful in various fields such as radiotherapy and basic research. It is important to produce separately between these nuclides depending on the application. In this work, the radioactivity ratios of ⁴⁶Sc and ⁴⁷Sc produced by photonuclear reactions from ^{nat}Ti and ⁴⁸Ti targets have been determined. ^{nat}TiO₂ and ⁴⁸TiO₂ were enclosed in a quartz tube for

bremsstrahlung radiation. The irradiations were carried out using the Electron Linear Accelerator at Tohoku University. The accelerator was operated at an electron energy of 20-50 MeV. The TiO₂ targets were placed in close contact with the back of a Ta converter and cooled with running tap water during the 10-30 minutes irradiation. After the irradiation, the targets were examined by gamma-spectrometry with a high purity Ge detector. As a result, the Sc-46/Sc-47 radioactivity ratios for ^{nat}Ti and ⁴⁸Ti targets were determined as a function of irradiation energy.

P37 Effect of Sodium Nitrate Concentration and pH on Adsorption Behavior of Trivalent Actinides and Lanthanides on Pyridine Resin in Aqueous Solution

FAUZIA, H.I.¹, ABE, C.², KONASHI, K.², SUZUKI, T.¹

(¹Nagaoka Univ. Technol., ²Tohoku Univ.)

Trivalent actinides (An (III)) and lanthanides (Ln) have similar ionic radii in aqueous solution. Thus, the separation of these elements is not so easy. We have been studying the adsorption behaviors of these elemental groups on pyridine resin. In our previous studies, we found that the group separation is available in HCl system, whereas in HNO₃ system, the group separation phenomenon does not occur as the adsorption is due to the ionic radii. The reason why this difference appears is that the adsorption mechanism of An(III) in HCl system is complex formation with pyridine group and the adsorption of An(III) and Ln in HNO₃ system is ion exchange. In the present study, we investigated the adsorption behavior of An(III) and Ln on pyridine resin in NaNO₃ aqueous solution with different concentration of NaNO₃ and pH of the aqueous solution. As a result, we found that the possibility of group separation of An(III) and Ln in HNO₃ system by suppressing the protonation of pyridine group.

P38 DFT calculation for elucidation of MA separation mechanism using C2-POPhen

NAKASHIMA, T.¹, FUKASAWA, Y.², NAKASHIMA, S.^{2,3}

(¹Grad. School of Sci., Hiroshima Univ., ²Grad. School of Sci. and Eng., Hiroshima Univ., ³N-BARD, Hiroshima Univ.)

Selective separation of minor actinides (MA) and lanthanides (Ln) is important in the treatment process of high-level radioactive liquid waste. The solvent extraction method is useful for the separation of MA and Ln and the elucidation of the separation mechanism using experiments and calculations is in progress. Recently, C2-POPhen (tetraethyl

(1,10-phenanthroline-2,9-diyl)phosphonate) has been studied as an extractant with high distribution ratio and MA selectivity[1]. In this study, we tried to clarify the extraction mechanism of C2-POPhen by using DFT (density functional theory) calculation. ML(NO₃)₃ and ML₂³⁺ (M=Eu³⁺, Am³⁺, L=C2-POPhen) models were created based on the similar complexes Y(CyMe₄-BTPPhen)(NO₃)(CN)₃ and Cu(C2-POPhen)₂PF₆Cl₆. Structural optimization and energy calculation were performed, and the bond order, molecular orbital, and MOOP (Molecular Orbital Overlap Population) were analyzed based on the energy results.

The magnitude of orbital interaction was evaluated using molecular orbital diagram and MOOP analysis. These results show that the overlap of Am's f orbital and coordinating N atom's orbital is larger than that of Eu's f orbital. From these results, it was clarified that the MA selectivity of C2-POPhen was affected by the difference in the metal f orbital and N atom orbital interactions.

[1] L. Xu et al., *Inorg. Chem.* 2019, **58**, 4420–4430

P39 An investigation on the laboratory management from the viewpoint of the management theory

TANIGUCHI, E.¹, ODA, H.²

(¹Chukyo Univ., ²Nagoya Univ.)

Most of the principal investigators (PI) of University are not well educated to be a laboratory manager; therefore, most laboratories have many management problems (Kwok, 2018). The purpose of this study is to clarify the typical problems on the management of chemical laboratory in Japanese graduate school and to indicate their countermeasures.

We, first, conducted unstructured interviews with principal investigators, other staffs and students about the problems of laboratory management. The interviews revealed the following three problems. The first is the phenomenon that students save their efforts to their researches while watching each other's progress. The second is that students do not voluntarily conduct the researches without their supervisor's guidance. The third is that students lose motivation for the current researches after obtaining a tentative job decision.

Next, we indicated their countermeasures from the viewpoint of the management theory. The first phenomenon is called "systematic soldiering". As the countermeasure, PI need to grasp the situation of students in detail. The second problem is called "flounder's paradox". The countermeasure is for PI to take leadership corresponding to student's maturity levels. The third is caused by a student's recognition that the purpose of their research is to obtain a master's degree. For this problem, PI should make students realize that current research will be useful for their future work.

P40 Tetrahedral Mesh Structure Modeling of New Generation Helical Fusion Reactor FFHR at NIFS for The Neutron Transport Simulation using a Monte-Carlo Based PHITS code

SAKAMA, M.¹, KOBAYASHI, M.², SAZE, T.², GOTO, T.², MIYAZAWA, J.², HAMABE, D.¹, KAMIYA, K.¹, YOKOTA, K.¹, OTANI, R.¹, MATSUMOTO-KAWAGUCHI, E.¹

(¹Tokushima Univ., ²NIFS)

Today, the National Institute for Fusion Science (NIFS) is energetically working on studies

relative to ultra high-temperature plasma and corresponding scientific phenomena on the state of matter for achieving a stable and permanent energy in future. Miyazawa's laboratory team (Fusion Engineering Research Project Team) in which we have collaborated from FY2019 is carrying out researches on New Generation Helical Fusion Reactor FFHR as the steady-state reactor replaced currently operating experimental-state reactor of LHD. In the research role of this team, we would like to develop the tetrahedral mesh structure modeling of FFHR and to simulate the neutron transport on the modeling calculation space when the FFHR will operate using PHITS code. In this presentation, we have investigated the tetrahedral mesh structure modeling associated with two paths toward the input code on PHITS what was done via .ele and .node files generated by TetGen program and the other was done via .bdf file generated by ANSYS Fluent.