

English Abstract

2S01 Bistable metal complexes

OSHIO, H

(Univ. of Tsukuba)

Molecules may have flexible structures with easily controlled electronic states. When molecules have several stable electronic states (or phases) and their structures are flexible upon state changes, such molecular systems can be promising candidates for molecular switching devices.

Spin crossover (SCO) and mixed valence (MV) complexes rest in two spin states and two valence states, respectively. Multi-component molecules, in which each component possesses several states due to spin conversion and electron transfer, are expected to show synergistic behaviors with stepped and/or selective state conversions. In this lecture, multi-component systems showing selective spin state conversion, photo-induced SCM, and lability controlled cluster syntheses will be presented.

2S02 Migration of radionuclides emitted from Fukushima Dai-ichi Nuclear Power Plant based on speciation and isotopic information

TAKAHASHI, Y.

(The University of Tokyo)

Speciation and isotope ratio of radionuclides contribute greatly to the better understanding of environmental behaviors of radionuclides emitted from Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in radiochemistry field. To reach this goal, development of hyper-sensitive speciation and isotopic ratio measurements for appropriate methods introduced from other fields is important, which can be coupled with conventional radioactivity measurement specific to this field. As examples of this strategy, development of X-ray absorption fine structure (XAFS) spectroscopy such as a hyper-sensitive XAFS method using wavelength detection mode with crystal analyzer and distribution analysis of functional groups in submicron scale using scanning transmission X-ray microscopy (STXM) will be given. In addition, systematic comparison of the results on radionuclides (e.g., cesium) with other elements enables us to predict element with little information on its environment behavior such as radium. As for the isotopic analysis, direct measurement of the isotopes in solid samples is very important (e.g., SIMS and La-ICPMS), if the available amount of the samples is very limited such as radiocesium-bearing particles (CsBP). The isotopic ratios of U and Cs showed that they are exclusively emitted from the FDNPP. Stable isotope fractionations of other elements such as Fe and Zn may help understand formation processes of CsBP including evaporation of these elements within FDNPP.

1A01 Observation of radiation situation in Iitate village during six years after the Fukushima-1 NPP accident and the current radiation level around living areas after decontamination

IMANAKA, T.¹, ENDO, S.², SUGAI, M.³, ICHIKAWA, K.³, HAYASHI, G.⁴, TOYODA, N.³, SAWAI, M.⁶, KAMISAWA, C.⁶, SAKUMA, J.³, ISHIDA, K.³, ITOU, N.³, BABA, H.³, ITONAGA, K.⁵, URAGAMI, K.⁵, OZAWA, S.³

(¹Kyoto Univ., ²Hiroshima Univ., ³IISORA, ⁴Tohoku Univ.,

⁵Nippon Univ. ⁶CNIC)

Car-borne radiation monitoring over the main roads of the Iitate village has been carried out periodically since the Fukushima-1 NPP accident in March 2011. Measured values inside the car were converted to those on the road. The average dose rate in the village was 10.7 $\mu\text{Sv/h}$ at the time of the first monitoring on March 29, 2011, while it decreased to 0.60 $\mu\text{Sv/h}$ at the eighth monitoring on April 1, 2017. A large scale decontamination work in Iitate village began in 2014 and continued for three years. Last year, in order to get the information on the current level of radiation and to prepare the planned lifting of evacuation instruction, radiation surveys around 249 houses (16 % of the total houses in village) were carried out in four districts in cooperation with local residents. The average levels of radiation were found to be about 0.5 – 1.0 $\mu\text{Sv/h}$. Assuming a house transmission factor of 0.4 and a stay model of 16-hr inside and 8-hr outside house, the annual radiation exposure of 3 – 5 mSv can be estimated when residents return to their homes.

1A02 Quantitative analysis of ³⁶Cl in surface soils near the Fukushima Dai-ichi Nuclear Power Plant using accelerator mass spectrometry

OHTA, Y.¹, SUEKI, K.¹, SASA, K.¹, TAKAHASHI, T.¹, MATSUNAKA, T.², MATSUMURA, M.¹, TOSAKI, Y.³, HOSOYA, S.¹, TAKANO, K.¹, OCHIAI, Y.¹, SATOU, Y.⁴, YOSHIKAWA, H.⁴, NAKAMA, S.⁴, MINOWA, H.⁵

(¹Univ. of Tsukuba, ²Kanazawa Univ., ³AIST, ⁴JAEA, ⁵Jikei Univ. School of Medicine)

A large amount of radionuclides was released into the environment by the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. The emission of ³⁶Cl was also suspected, whereas only one report so far measured ³⁶Cl in contaminated soil. Therefore, we conducted additional measurements of ³⁶Cl in soil around the FDNPP. We collected surface 5 cm soil at 31 locations around the FDNPP (distance from the FDNPP: 1.4-28.8 km) and extracted inorganic chlorine (Cl_{inorg}) from the soil by 0.01 M HNO_3 . The ³⁶Cl/Cl was determined by AMS at the University of Tsukuba, Tandem Accelerator Complex. In 10 locations among the 31 locations (distance from the FDNPP: 7.6-28.8 km), the ³⁶Cl/Cl varied from 0.138 to 6.86 ($\times 10^{-12}$) and ³⁶Cl inventory varied from 0.132 to 3.96 mBq/m². Some of the measured ³⁶Cl/Cl were higher than those before the accident. In these sampling points, the observed ³⁶Cl inventory were also larger than the estimated value before the accident. These results are considered to be evidence of the ³⁶Cl emission. However, the definite correlation was not found between ³⁶Cl and ¹²⁹I or ¹³⁷Cs. The sampling points with larger ³⁶Cl inventory were located at a distance of 7.56-8.12 km from the FDNPP. Measurement results in another 21 locations including those closer to the FDNPP will be shown in the presentation.

1A03 Radioactivity monitoring in the Tokyo Bay – temporal variations of ¹³⁴Cs and ¹³⁷Cs in sea sediment and seawater –

TOYOOKA, S.¹, NITTA, W.¹, KANEKO, K.¹, KISHIMOTO, T.¹, ISOGAI, K.¹,

(¹ Japan Chemical Analysis Center)

After the accident of Fukushima Dai-ichi NPP, a large amount of

radioactive materials had been released into the environment and distributed widely in the Kanto area. Atmospheric radioactive materials deposited on the ground surface by rainfall, were transported by natural phenomenon or anthropogenic action. And finally flowed into the Tokyo Bay via the rivers, and deposited on sediment.

Radioactivity monitoring of ^{134}Cs and ^{137}Cs at 23 points in the Tokyo Bay has been carried out since 2013 to estimate the influence of Fukushima Dai-ichi NPP accident. In recent four years, activity concentrations has been decreased at most points. As for, point “K-T1” and “C-P8” which are located off Arakawa River and Kyu-Edogawa River, and near Kisarazu Port, activity concentrations maintained relatively high during this monitoring.

Activity concentrations of “K-T1” and “M-C1” which are located off Arakawa River and Kyu-Edogawa River, showed a different secular variation in spite of the same river mouth. Although “K-T1” is farther than “M-C1” from the river mouth, radioactivity concentrations in “K-T1” is higher than that in “M-C1”.

Radioactivity ratios of $^{134}\text{Cs}/^{137}\text{Cs}$ show that the source term of radio cesium in sediment and seawater samples had been derived from the accident of Fukushima Dai-ichi NPP.

1A04 Existence forms and distribution characteristics of radiocesium in coastal marine sediments off Fukushima Prefecture

TERASAKI, S.¹, NAGAO, S.¹, ODANO, N.², ASAMI, M.²
(¹LLRL, Kanazawa Univ., ²National Maritime Research Institute)

Radiocesium was released into marine environment because of the accident of Fukushima Dai-ichi Nuclear Power Plant (FDNPP). A part of radiocesium was adsorbed to the bottom sediments and their radiocesium concentration varied spatially and temporally. This study investigates the distribution characteristics of ^{137}Cs and the existence forms by using selective extraction experiments. Sediment samples were collected in the coastal area off Fukushima and Miyagi Prefectures in 2014 and 2015. Gamma rays of ^{134}Cs (605 and 796 keV) and ^{137}Cs (662 keV) were measured for the samples. Artificial seawater dissolution experiment and selective extraction experiments (1M ammonium acetate and 10% hydrogen peroxide) were carried out for the sediment samples. Water content is related to the variations of radiocesium concentration. Dissolution rate of ^{137}Cs by artificial seawater ranges from 0.047 to 0.16% for the sediments off the Abukuma River and from 0.38 to 14% for those off the Niida River and FDNPP. The results indicate that most of radiocesium is adsorbed to clay minerals in the sediments and the variation of ^{137}Cs is controlled by the movement of sediment particles.

1A05 Research for the distribution of radiocesium-bearing particles in dust from near the Fukushima Daiichi Nuclear Power Plant

MINOWA, H.¹, YOSHIKAWA, H.², NAKAMA, S.², SATOU, Y.², SUEKI, K.³
(¹Jikei Univ., ²JAEA, ³Univ. of Tsukuba)

Radiocesium-bearing particles in dust samples from 27 location were investigated from January 2014 to November 2016,

at Futaba-machi and Okuma-machi within about 8 km from Fukushima Daiichi Nuclear Power Plant. Dust samples were collected by an adhesive sheet or a smear filter paper from the smooth surface within 10×10 cm in the place where the air from the outdoor enters and not exposed to rain, such as a garage, a storehouse or under the ventilation fan in the kitchen. Radiocesium-bearing particles were picked up from the dust collected with a spatula or a brush. Particulate spots were observed in the image of autoradiography for the adhesive sheets. The size and the concentration (indicator of the radioactivity) and the number of the particulate spots were greatly different depending on the sampling place. The smear filter samples were measured by the γ -ray spectrometry with a Ge-detector. Based on the quantitative ^{134}Cs and ^{137}Cs values, the $^{134}\text{Cs}/^{137}\text{Cs}$ ratios at March 2011 were calculated, and the origin reactor and its mixing ratios were estimated. The particles were analyzed by SEM and their shape observation and chemical composition were determined. Based on these data, the spatial distribution of radiocesium-bearing particles and the origin of particles and their transportation/deposition processes will be discussed.

1A06 Production experiments of the insoluble Cs-concentrated particles including metal elements released by the FDNPP accident

FUJITA, N.¹, NINOMIYA, K.¹, SHINOHARA, A.¹, YOSHIMURA, T.², KOGURE, T.³

(¹Graduate School of Science, Osaka University, ²Radioisotope Research Center, Osaka University, ³Graduate School of Science, The University of Tokyo)

By the accident of Fukushima Daiichi Nuclear Power Plant (FDNPP), many amounts of radioactive nuclides were released into the environment. Among them, the existence of Cs-concentrated particles with a diameter of about 2 μm was reported. The main component of the particle is SiO_2 , and it also contains Cs, Fe, and Zn. The chemical property of the particle is glassy and insoluble in water and acid. In this study, we produced the simulated particles by generating a splash of SiO_2 in melting CsOH in 300~500 $^\circ\text{C}$ temperature condition. We also produced particles containing Fe and Zn by putting galvanized iron piece in melting CsOH . We observed the produced particles using Scanning Electron Microscope (SEM), Energy Dispersive X-ray Spectroscopy (EDS), and Transmission Electron Microscope (TEM). We found that the particles have the similar shape and internal composition with the observed insoluble Cs-concentrated particles.

1A07 Radioactive cesium-bearing particles in black substances

KURIHARA, Y.¹, MIURA, H.¹, HIGAKI, S.², SAKAGUCHI, A.³, TANAKA, K.⁴, NAGAO, S.⁵, YAMAMOTO, M.⁶, TAKAHASHI, Y.¹

(¹The Univ. of Tokyo, ²RIC, The Univ. of Tokyo, ³Univ. of Tsukuba, ⁴JAEA, ⁵LLRL, Kanazawa Univ., ⁶Kanazawa Univ.)

In order to obtain the comprehensive information on the characteristics of the radioactive Cs-bearing particles (RCs particles) and their distribution in Fukushima contaminated areas,

the RCs particles in the black substances (highly contaminated road dust) were investigated. The activities of ^{134}Cs and ^{137}Cs in the particles separated from the sample and identified by SEM-EDS were measured by HPGe detector. Spherical particles with diameters of 5 μm or less were found in any sampling areas. Particles of this type are similar in terms of chemical composition to those reported so far, and were estimated to be derived from Units 2 or 3 of FDNPP from the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio. In the samples of Futaba Town and Minamisoma City, particles with diameters of several tens to several hundreds of microns were found. Most of the particles in this region were not uniform in shape. The main components of the particles were Si, Ca, K, and Al, which were estimated to be derived from Unit 1 of FDNPP from the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio. From the results above, it was suggested that large and small particles have different sources and generation processes. Additionally, it was suggested that their generation processes are different even among small particles, due to the difference in the composition and the relationship between ^{137}Cs radioactivity and the volume.

1A08 Monthly flux of particulate ^{129}I in river of Fukushima from 2013 to 2016

MATSUNAKA, T.^{1,2}, SASA, K.¹, TAKAHASHI, T.¹, ONDA, Y.¹, TANIGUCHI, K.³, WAKIYAMA, Y.⁴, SUEKI, K.¹
(¹Univ. of Tsukuba, ²Kanazawa Univ., ³Fukushima Pref. Gav., ⁴Fukushima Univ.)

This study investigated the fluxes of particulate ^{129}I and ^{137}Cs in the downstream of Niida River system in Fukushima, where the upper watershed is in the relatively high-contaminated zone located 30–40 km northwest from the Fukushima Dai-ichi Nuclear Power Plant. Suspended substance (SS) in river water were continuously collected at Haramachi site (5.5 km upstream from the river mouth) of the downstream river by installing the time-integrative SS sampler from Jan. 2013 to Jun. 2016. Monthly activities of ^{129}I and ^{137}Cs in SS were 0.92–4.1 mBq kg⁻¹ and 16–31 kBq kg⁻¹ during the 2013. The ^{129}I activity was strongly correlated with the maximum turbidity in the river water with correlation coefficient (R^2) of 0.90. Higher ^{129}I activity found in Mar. Apr., Sep., and Oct. 2013 were considered to infer the high contribution of SS from higher contaminated upper watershed area. Meanwhile, the fluxes of particulate ^{129}I and ^{137}Cs at Haramachi site were estimated to be 0.024–43 kBq month⁻¹ and 0.70–330 GBq month⁻¹ from 2013 to 2016. There is a possibility that relatively massive amount of particulate ^{129}I and ^{137}Cs from the upstream were transported to the downstream of the river and marine environment by the rain events.

1A09 Transport behavior of particulate forms of radiocesium observed at sediment trap experiments off the Fukushima Dai-ichi Nuclear Power Plant

NAGAO, S.¹, TERASAKI, S.¹, ODANO, N.², ASAMI, M.²
(¹KANAZAWA Univ., ²NMRI)

The sediment trap experiments were carried out at two sites of the coastal area off the Fukushima Dai-ichi Nuclear Power Plant. The total mass flux of settling particles and radiocesium radioactivity were monitored at a 30 m water depth site in a period

of 6 October–6 November 2015 and 26 December 2015–26 January 2016, and at a 60 m water depth site during 6 October–6 November 2015. Higher total mass flux and radiocesium flux were observed for the settling samples collected at both sites during stormy weather condition (6–11 October 2015). The similar situation was also observed during 20–26 January 2016. The settling particles were sandy materials with average particle size of 33–133 μm . The results indicate that sandy bottom sediments were re-suspended and moved in the coastal area less than water depth of 60 m during stormy weather condition.

1A10 The transport of radiocesium in sediments by flow of river at Tamagawa river watershed

AKIYAMA, M.¹, OKUMURA, S.¹, SHIMONO, A.², OCHI, K.³, HAGIWARA, K.², KOIKE, Y.²

(¹ Grad. Sch. of Sci. and Tech., Meiji Univ., ² Sch. of Sci. and Tech., Meiji Univ., ³ JAEA)

Radionuclides including radiocesium (^{134}Cs and ^{137}Cs) was spread out from the Fukushima Daiichi Nuclear Power Plant (FDNPP) after the accident. The behavior of FDNPP-derived radiocesium in sediment were observed at 50 sampling points in Tamagawa river watershed. The Tamagawa river watershed is an urban river flowing throughout the Tokyo Metropolis. The activity concentrations of radiocesium in sediment at downstream were higher than that at upstream. On the other hand, high concentration of radiocesium in sediment was observed at upstream of Nogawa river. The flow rate of Nogawa river is slower than that of Tamagawa river watershed because Nogawa river is one of the tributary of Tamagawa river watershed. These results indicated that the transport process of radiocesium in sediment depend on the water discharge and watershed environment.

1A11 The effect of radiocesium-bearing microparticles on the solid-water distribution coefficient of radiocesium in rivers in Fukushima

*Hikaru Miura¹, Yuichi Kurihara¹, Aya Sakaguchi², Seiya Nagao³, Kazuya Tanaka⁴, Shogo Higaki¹ and Yoshio Takahashi¹

(¹Univ. of Tokyo, ²Univ. of Tsukuba, ³Univ. of Kanazawa, ⁴JAEA)

We focused on the radiocesium-bearing microparticles (CsBPs) in suspended particles which were collected from rivers in Fukushima by filtering water in 2011–2016. Radiocesium in CsBP incorporated in the SiO₂ glass is generally water-insoluble. Thus, cesium (Cs) in CsBP is not likely involved in the adsorption-desorption reaction. According to Konoplev et al. (2016), (i) K_d in rivers in Fukushima are higher than those in Chernobyl by 1–2 orders of magnitude, and (ii) it is partly because radiocesium-bearing microparticles (CsBPs) are present in solid materials in Fukushima area. We calculated the ratio of ^{137}Cs in CsBPs to the total ^{137}Cs in the suspended particles to know the effect of CsBPs on K_d in rivers. The results of SEM-EDS showed that the size, shape, and chemical compositions of CsBPs were similar to those reported as spherical CsBPs. The radioactivity ratio of CsBPs to total ^{137}Cs on the filters ranged from 1.4–46%, which makes $\log K_d$ value apparently higher by only 0.27. This result means that the higher K_d in Fukushima than in Chernobyl could not be explained only by the presence of CsBPs. The fact

that the presence of high concentration of organic matter in Chernobyl inhibits strong association of RCs with phyllosilicate minerals should be considered, which can make K_d lower in Chernobyl.

1B01 Measurement of the half-lives and the internal-conversion electron energy spectra for several chemical forms of ^{235m}U

SHIGEKAWA, Y.¹, KASAMATSU, Y.¹, YASUDA, Y.¹, KANEKO, M.², SHINOHARA, A.¹

(¹Grad. Sch. Sci., Osaka Univ., ²JAEA)

The nuclide ^{235m}U is the isomeric state of ^{235}U with extremely low excitation energy (76.8 eV), and decays through the internal conversion (IC) process. Because outer-shell electrons are dominantly involved in the IC process of ^{235m}U , it is indeed reported that the decay constant (half-life) of the isomer varies depending on its chemical environment. The half-life variation does not show simple tendency for some chemical forms of ^{235m}U , and the mechanism is not understood well. In order to elucidate the deexcitation process of ^{235m}U , we are trying to measure the energy spectra of IC electrons in addition to the half-lives for various chemical environments. In this work, we collected ^{235m}U recoiling out of a ^{239}Pu source on Cu sheets by using Collection Apparatus for Recoil Products (CARP), and we measured the half-lives and the energy spectra using a retarding-field type spectrometer. By the reaction of ^{235m}U with several gases (HF, HCl, O₂), we prepared some kinds of chemical forms of ^{235m}U and measured their half-lives and energy spectra. Consequently, we observed clear difference in half-life and energy spectrum between ^{235m}U on Cu and ^{235m}U reacted with HF gas. We performed relativistic DFT calculation to discuss the relationship between the electron orbitals and the half-lives of ^{235m}U .

1B02 Measurement of total kinetic energy of fission fragments in SF decay of ^{254}Rf

GOTO, S.¹, KAJI, D.², TSUCHIYA, S.¹, AONO, R.¹, MORIMOTO, K.², HABA, H.², OOE, K.¹, KUDO, H.¹

(¹Niigata Univ., ²Nishina Center, RIKEN)

We are planning to systematically measure properties of spontaneous fission of rutherfordium isotopes systematically, such as mass and total kinetic energy (TKE) distributions of fission fragments, in order to investigate the nature of SF of superheavy nuclei in detail. Aono *et al.* observed SF events of $^{253,254}\text{Rf}$ in $^{208}\text{Pb} + ^{48}\text{Ti}$ reaction and measured TKEs in SF decay of ^{254}Rf . The results showed that TKEs in ^{254}Rf indicated a rather different trend expected from those in ^{256}Rf . However, the observed TKE has much uncertainty because of poor statistics. In this work, TKE in SF decay of ^{254}Rf was measured with better statistics in $^{206}\text{Pb} + ^{50}\text{Ti}$ reaction.

Experiments were performed at the RIKEN linear accelerator (RILAC) facility using a gas-filled recoil ion separator, GARIS. ^{254}Rf was produced by $^{206}\text{Pb}(^{50}\text{Ti}, 2n)$ reaction. ^{256}Rf was also produced by $^{208}\text{Pb}(^{50}\text{Ti}, 2n)$ reaction. ^{206}Pb and ^{208}Pb targets of $\sim 350 \mu\text{g cm}^{-2}$ and $\sim 400 \mu\text{g cm}^{-2}$ in thickness, respectively, were prepared by a vacuum evaporation on 60 $\mu\text{g cm}^{-2}$ carbon foils. Beam energies of ^{50}Ti were 241 MeV for ^{206}Pb and 239 MeV for

^{208}Pb . Average intensity of ^{50}Ti was 280 pnA.

A total of 740 events was observed as SF decay of ^{256}Rf in $^{206}\text{Pb} + ^{50}\text{Ti}$ reaction. Deduced half-life is 6.30 ± 0.22 ms, which agrees with the previously reported value. A preliminary analysis shows that the half life of ^{254}Rf is about 28 μs which agrees with the reported value. In the presentation, we will report the results of analysis in detail.

1B03 Production of isotopes of element 107, Bh in the $^{248}\text{Cm} + ^{23}\text{Na}$ reaction and their decay studies for chemistry studies of Bh

KOMORI, Y.¹, HABA, H.¹, FAN, F.², KAJI, D.¹, KASAMATSU, Y.³, KIKUNAGA, H.⁴, KONDO, N.³, KUDO, H.^{1,5}, MORIMOTO, K.¹, MORITA, K.^{1,6}, MURAKAMI, M.¹, NISHIO, K.⁷, OMTVEDT, J. P.⁸, OOE, K.⁵, QIN, Z.², SATO, D.⁵, SATO, N.¹, SATO, T., SHIGEKAWA, Y.³, SHINOHARA, A.³, TAKEYAMA, M.^{1,9}, TANAKA, T.^{1,6}, TOYOSHIMA, A.⁷, TSUKADA, K.⁷, WAKABAYASHI, Y.¹, WANG, Y.², WULFF, S.⁸, YAMAKI, S.¹, YANO, S.¹, YASUDA, Y.³, YOKOKITA, T.¹

(¹RIKEN, ²IMP, ³Osaka Univ., ⁴Tohoku Univ., ⁵Niigata Univ., ⁶Kyushu Univ., ⁷JAEA, ⁸Univ. Oslo, ⁹Yamagata Univ.)

We plan to study chemical properties of element 107, Bh using the gas-filled recoil ion separator (GARIS) at the RIKEN linear accelerator facility. In this work, we investigated production cross sections of Bh isotopes in the $^{248}\text{Cm} + ^{23}\text{Na}$ reactions and their decay properties such as α -particle energy and half-life. The Bh isotopes were produced in the $^{248}\text{Cm}(^{23}\text{Na},xn)^{271-x}\text{Bh}$ reactions at the ^{23}Na beam energies of $E_{\text{lab}} = 121, 126, 131, \text{ and } 135$ MeV. The evaporation residues were separated in flight from the beam particles and nuclear transfer reaction products by GARIS and were extracted to the chemistry laboratory by the He/KCl gas-jet system. Alpha particles and SF fragments were measured with the rotating wheel system MANON for α /SF-spectrometry. We observed time-correlated 11 α - α and 12 α -SF events, and they were reasonably assigned to ^{266}Bh and its α -decay daughter ^{262}Db and granddaughter ^{258}Lr . No clear α - α correlations on ^{265}Bh and ^{267}Bh were found. The α -particle energies of ^{266}Bh were observed in the wide range of $E_{\alpha} = 8.62\text{--}9.40$ MeV and the half-life of ^{266}Bh was determined to be $T_{1/2} = 10.0^{+2.6}_{-1.7}$ s. This half-life is about 8 times longer than the literature values for ^{266}Bh . The excitation function for the $^{248}\text{Cm}(^{23}\text{Na},5n)^{266}\text{Bh}$ reaction shows the maximum cross section of $\sigma = 57 \pm 13$ pb at $E_{\text{lab}} = 131$ MeV.

1B04 High-precision mass measurements of short-lived nuclei with MRTOF+ GARIS-II

NIWASE, T.^{1,2}, WADA, M.^{1,3}, SCHURY, P.³, ITO, Y.^{1,8}, KIMURA, S.^{1,4}, ROSENBUSCH, M.¹, KAJI, D.¹, MORIMOTO, K.¹, HABA, H.¹, YAMAKI, S.^{1,5}, TANAKA, T.^{1,2}, MORITA, K.^{1,2}, TAKAMINE, A.¹, MIYATAKE, H.³, HIRAYAMA, Y.³, WATANABE, Y.³, MOON, J. Y.^{3,6}, MUKAI, M.⁴, WOLLNIK, H.^{1,7}, (¹RIKEN Nishina Center, ²Kyushu Univ., ³KEK, ⁴Univ. of Tsukuba, ⁵Saitama Univ., ⁶IBS, ⁷NMSU, ⁸McGill Univ.)

The binding energy is one of the basic physical quantities, that is related to stability of nuclei and their decay properties. It can be derived from measuring the mass precisely. A multi-reflection time-of-flight mass spectrograph (MRTOF-MS) coupled with a

gas-filled recoil ion separator (GARIS-II) is a promising apparatus for measuring the mass of superheavy nuclei directly. Recently, we succeeded to measure the mass of several-tens of heavy-nuclides, which were produced via heavy-ion induced fusion reactions, using MRTOF-MS. The system was applicable to short-lived nuclides with a few-tens of milliseconds. In this presentation, we will report their results in detail. In addition, we will also talk about the development of a new α -ToF detector, which enables to measure both mass of superheavy nucleus and alpha-decay related with it.

1B05 Production of Tc isotopes for Compton Camera(ETCC) imaging

HATSUKAWA, Y.¹, TSUKADA, K.², HASHIMOTO, K.¹, SATO, T.², ASAI, M.², TOYOSHIMA, A.², NAGAI, Y.¹, TANIMORI, T.³, SONODA, S.³, KABUKI, S.⁴, KIMURA, H.⁵, TAKADA, A.³, MIZUMOTO, T.³, HAYAKAWA, T.¹, Takaki, S.²
(¹QST, ²JAEA, ³Kyoto Univ., ⁴Tokai Univ., ⁵Kyoto Pharm. Univ.)

In recent years, the Compton camera which is originally developed for the astrophysical studies was applied for medical diagnostic usage. For the Compton camera imaging, isotopes emitting higher energy gamma-rays were required. If technetium isotope which emits higher energy gamma-rays is available for synthesizing labelled compound, All of labelled compound which is previously developed for Tc-99m can be available for Compton camera imaging using other Tc isotope. Two Tc isotopes, ^{95m}Tc ($T_{1/2} = 60$ d; $E_{\gamma} = 204, 582$ and 835 keV) and ⁹⁶Tc ($T_{1/2} = 4.28$ d, $E_{\gamma} = 778$ and 812 keV) are strong candidates for this purpose. These isotopes were produced by proton irradiation with enriched molybdenum trioxide target. In this study, recycling of enriched molybdenum targets was developed. Identification of obtained material was examined by X-ray diffraction, XRD method. Recovery yield of molybdenum trioxide obtained by this recycling method are 70-90 %.

1B06 Synthesis of submicron-sized calcium molybdates, using liposome as a template, and characterization of their physicochemical properties

KURITA, S.¹, YAMASAKI, S.¹, OCHIAI, A.², UTSUNOMIYA, S.², SAKAGUCHI, A.¹, SUEKI, K.¹
(¹Univ. of Tsukuba, ²Kyushu Univ.)

Technetium-99m has been commonly used for a nuclear medicine diagnosis and ^{99m}Tc is separated from ⁹⁹Mo. Numerous ⁹⁹Mo production methods, such as ¹⁰⁰Mo(n, 2n)⁹⁹Mo reaction, have been investigated. After the reaction, ⁹⁹Mo and ^{99m}Tc have been separated from each other by using sublimation method. Then, we aim to make the method more simplified by preparing the ¹⁰⁰Mo material, which enable us to recover only ^{99m}Tc from the material at operation temperature. We propose submicron calcium molybdate is better material. In the present study, we focused on the synthesis method of submicron calcium molybdate using liposome as a template. Liposome has been used for synthesis of biocompatible materials such as calcium carbonate and calcium phosphate nanoparticle. We applied the synthesis procedure using liposome to the calcium molybdate formation. The calcium molybdate particles prepared in this study were characterized by

the electron microscopy, zeta potential measurement and so on, to obtain the physicochemical properties such as particle size, composition, morphology and crystalline. The products, which we can get under present experiment condition, were identified to be calcium molybdate by the SEM and TEM observation. We discuss about the properties of the products and the template effect of liposome on the calcium molybdate formation.

1B07 Production of purified ^{135m}Ba for fee-based distribution using the ¹³³Cs(α, pn)^{135m}Ba reaction

YANO, S.¹, HABA, H.¹, KOMORI, Y.¹, YOKOKITA, T.¹, TAKAHASHI, K.¹, SATO, N.¹, OSANAI, M.¹
(¹RIKEN Nishina Center)

Barium-135m ($T_{1/2} = 28.7$ h, $E_{\gamma} = 268.2$ keV) is one of the useful radiotracers of Ba in various research fields such as nuclear medicine. In this work, we investigated a production method of a purified ^{135m}Ba for a fee-based distribution to general public in Japan.

Barium-135m was produced in the ¹³³Cs(α, pn)^{135m}Ba reaction by bombarding a CsCl target with a 50.5-MeV alpha beam from the RIKEN AVF cyclotron. After the irradiation, ^{135m}Ba was purified by the reversed-phase extraction chromatography using the Eichrom Sr resin. We evaluated a thick-target yield and a radionuclidic purity of the purified ^{135m}Ba via γ -ray spectrometry.

The thick-target yield of ^{135m}Ba was determined to be 2.1 ± 0.1 MBq $\mu\text{A}^{-1} \text{h}^{-1}$ at 50.5 MeV. This yield is smaller by a factor of 4 than that reported by Dmitriev and Panarin in 1979 at 43.5 MeV. The chemical yield of ^{135m}Ba was about 80%. The radionuclidic purity of the purified ^{135m}Ba was about 70% due to byproducts of ^{133m}Ba and ^{133g}Ba. In the symposium, the specific radioactivity and the chemical impurity of the purified ^{135m}Ba will be also reported after a chemical element analysis with ICP-MS.

1B08 Development of a ²¹¹Rn/²¹¹At Generator Based on Dry-Chemistry

NISHINAKA, I.¹, HASHIMOTO, K.¹, MAKII, H.²
(¹QuBS, QST, ²ASRC, JAEA)

Astatine-211 is a prospective candidate for target alpha therapy. Our project focuses on a ²¹¹Rn/²¹¹At generator. The daughter ²¹¹At of 7.2 h in half-life is generated through EC decay of the parent ²¹¹Rn of 14.7 h, expanding time-frame for transportation and use of ²¹¹At. In this project, a chemical procedure based on dry-chemistry has been studied to develop the generator system.

Radon-211 was produced in the irradiation of a thin bismuth target with 60 MeV ⁷Li³⁺ beams from the JAEA tandem accelerator via the ²⁰⁹Bi(⁷Li, 5n)²¹¹Rn reaction. After the irradiation, the ²¹¹Rn was separated from the target and purified in a chemical procedure based on dry-chemistry using an apparatus for the generator system. The ²¹¹Rn in the apparatus was monitored by lead-shielded 1cm³ CdZnTe solid state detectors. After a half day, ²¹¹At generated from the purified ²¹¹Rn was eluted with ethanol.

The overall recovery yields of ²¹¹At generated from ²¹¹Rn (n=3) were 35% in the first fraction of the eluted ethanol of 2 mL, 8% in the second and 1% in the third. The results demonstrate that the chemical procedure based on dry-chemistry could provide the unique ability of the ²¹¹Rn/²¹¹At generator.

1B09 Extraction behavior of Astatine into hydrocarbon solvents

WASHIYAMA, K.¹, SHIN, Y.², YOKOYAMA, A.², TOYOSHIMA, A.^{3,4}, KANDA, A.⁴, SHINOHARA, A.⁴, YOSHIMURA, T.⁵

(¹Institute of Med. Pharm. and Health Sci. Kanazawa Univ., ²College and Institute of Sci. and Eng., Kanazawa Univ., ³Advanced Sci. Res. Center, JAEA, ⁴Dep. Of Chem., Graduate School of Sci., Osaka Univ., ⁵Radioisotope Res. Center, Osaka Univ.)

Since astatine-211 (²¹¹At) releases alpha particles with a moderate half-life (7.21 h), it is expected to be applied to radionuclide therapy. To obtain a high radioactivity of ²¹¹At after target irradiation, rapid and efficient separation method are warranted. To upgrade the wet chemistry, it is important to establish an optimal solvent extraction system considering the process after separation from the target, but no definitive ones have been reported. In this study, we evaluated the extraction behavior of astatine with respect to several hydrocarbon solvents as another candidate of the extraction solvents.

The hydrocarbon solvents used were hexane, octane, decane, dodecane, tetradecane, hexadecane, and some structural isomers of hexane. After solvent extraction of ²¹¹At into hydrocarbons from a nitric acid, we determined the distribution ratio (*D*) as the concentration ratio of ²¹¹At in the aqueous phase and organic phase.

The *D* values of ²¹¹At into dodecane was increased with increasing shaking time and reached an equilibrium state after 10 minutes. The *D* values of ²¹¹At into each hydrocarbon solvent with 1M HNO₃ increased with molecular weight. In addition, although hexane showed a low *D* value, all other structural isomers showed high *D* values. These imply that the ²¹¹At extraction correlates not only to dipole momentum but also to the polarization of each solvent.

1B10 Development of a simplified synthesis method for ²¹¹At-labeled medicine using resin-supported bifunctional reagent

KANDA, A.¹, TOYOSHIMA, A.^{2,3}, IKEDA, A.¹, ICHIMURA, S.⁴, ZHANG, Z.^{1,3}, YOSHIMURA, T.^{3,5}, SHINOHARA, A.^{1,3}.

(¹Grad. Sch. of Sci., Osaka Univ., ²Advanced Sci. Res. Center, Japan Atomic Energy Agency, ³ Project Res. Center for Fundamental Sci., Grad. Sch. of Sci., Osaka Univ., ⁴Fac. of Sci., Osaka Univ., ⁵Radioisotope Center, Osaka Univ.)

Recently, we aim to develop a new automated ²¹¹At separation and labeling method by seamless connection of column-flow labeling procedure to a dry distillation process. So far, we have developed a new resin, which can hold At and release it to medicine, used in the column flow method and then performed batch-wise labeling experiments. In this work, we carried out to label proteins with ²¹¹At in column-flow experiments. Injected ²¹¹At was first held on the resin under an oxidative condition with N-iodosuccinimide. After quenching excess trimethylstannyl residue and the oxidant, resin was washed with MeOH/1% AcOH. Then, proteins in PBS-T were eluted to react with the resin. The protein effluent was then ultra-filtrated using a membrane filter.

Control experiments without proteins were also carried out. As a result, radioactivity yields of the labeled protein in the supernatant was 4.3%, while that of the blank is 0.5%. This result demonstrated successful ²¹¹At labeling of proteins by the column-flow method. We will also present a column-flow labeling experiment of ammine in the symposium.

1B11 Developments of quantitative imaging system for alpha-emitting radioisotope ²¹¹At

SEGAWA, M.¹, NISHINAKA, I.², INOUE, T.³, TOH, Y.¹

(¹Japan Atomic Energy Agency, ²National Institute for Quantum and Radiological Science and Technology, ³Photron)

Radio nuclide therapy using α -particle emitted from ²¹¹At is of current interest because α -particles have higher LET. The establishment of effective technique to generate ²¹¹At has been recently required. Here, real-time imaging techniques to analyze the ²¹¹At and other radioactive materials generated at the same time will be great help for the establishment of effective techniques. Thus, we have developed two real-time camera systems adjusted for high resolution imaging and high sensitive imaging for relatively low-activity α -particles of ²¹¹At in the research phase. ²¹¹At was generated by the ²⁰⁹Bi (⁷Li, 5n) ²¹¹Rn \rightarrow ²¹¹At reaction at Japan Atomic Energy Agency. In this presentation, the recent results for the two systems described above are reported.

2A01 Rapid determination of ⁹⁰Sr in urine samples by extraction chromatography coupled to ICP-MS/MS

TOMITA, J.¹, TAKEUCHI, E.¹

(¹JAEA)

A rapid analytical method of ⁹⁰Sr in urine samples by extraction chromatography coupled to triple-quadrupole inductively coupled plasma-mass spectrometry (ICP-MS/MS) was examined. Strontium in urine sample was purified by phosphate coprecipitation followed by extraction chromatography. To eliminate both isobaric (⁹⁰Zr) and polyatomic (Ge, Se) interference for ⁹⁰Sr measurement by ICP-MS/MS, Sr was separated with tandem of Pre-filter, TRU and Sr resin. The measurement of 0.02 M HNO₃ solution with 50 mg/L of Sr indicated that tailing of ⁸⁸Sr does not impact the measurement of ⁹⁰Sr at concentrations up to 50 mg/L of Sr. Stable Sr and ⁹⁰Sr contents in the elution were measured by high abundance sensitivity ICP-MS/MS with O₂ gas reaction mode. The validation of this analytical method was confirmed by the analyses of synthesis urine sample containing the known amount of ⁹⁰Sr along with 1 mg of Ge, Se, Sr and Zr. The detection limit of ⁹⁰Sr was achieved to be approximately 1.0 Bq per urine sample, and analysis time needed for separation of Sr and ⁹⁰Sr measurement by ICP-MS was about 10 hours.

2A02 Trial measurements of ⁹⁰Sr with accelerator mass spectrometry

HOSOYA, S.¹, SASA, K.¹, TAKAHASHI, T.¹, HONDA, M.¹, SATOU, Y.², TAKANO, K.¹, OCHIAI, Y.¹, YAMAGUCHI, K.¹, SUEKI, K.¹

(¹Univ. of Tsukuba, ²JAEA.)

Strontium-90 (^{90}Sr , $T_{1/2} = 28.79$ yr) is an important radioactive nuclide that released into the environment from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. Common analytical techniques take too much time to analyze ^{90}Sr such as beta-counting. Recently, the new method with the ICP-MS was developed for rapid ^{90}Sr measurements. However it lacks sufficient technical capacity to assay ^{90}Sr pollution status for the FDNPP accident. If the ^{90}Sr measurement becomes possible with accelerator mass spectrometry (AMS), it will be available for the rapidly and high sensitive ^{90}Sr measurements. There are difficult problems to be solved for ^{90}Sr AMS in regard to the low beam current and the isobaric interference of ^{90}Zr . We prepared SrF_2 sample mixed with PbF_2 powder for high current beams of SrF_3^- . Consequently $^{88}\text{SrF}_3^-$ beam current was obtained about 300 nA. However, we couldn't identify ^{90}Sr spectrum, because there were too many ^{90}Zr in PbF_2 powder. Therefore we have to reconsider about materials of mixed powder. Standard samples were prepared from IAEA-PT samples ($^{90}\text{Sr} = 14.7 \pm 0.5, 29.6 \pm 0.8$ Bq/kg). However standard samples need to remove Zr by chemical methods because they contain Zr at ~ 7 ppt. In the future, we have a plan to modify the ion source to achieve the maximum beam current of 500 nA.

2A03 Speciation analysis of ^{129}I from the Fukushima Dai-ichi Nuclear Power Plant accident in natural samples

HONDA, M.¹, TAKAKU, Y.², MATSUZAKI, H.³, SASA, K.¹, SUEKI, K.¹

(¹Univ. of Tsukuba, ²Institute for Environmental Science, ³The University of Tokyo)

Iodine-129 (^{129}I : $t_{1/2} = 15.7$ Myr) has been introduced into the terrestrial environment through spent nuclear fuel reprocessing operations and nuclear accidents. The regional $^{129}\text{I}/^{127}\text{I}$ ratio in Japan was significantly increased by the Fukushima Dai-ichi nuclear power plant accident. The study of different iodine species, colloidal iodine and ionic iodine, is essential because its subsurface transport, fate as well as bioavailability strongly depend on its physicochemical forms (species). The $^{129}\text{I}/^{127}\text{I}$ and iodine amounts of individual chemical species were identified for pond water from Fukushima and riverine water from Chiba, Japan. Analytical results revealed that colloidal ^{129}I was the dominant species and accounted for more than 90% of the total ^{129}I present, whereas ionic ^{127}I was the dominant species and accounted for 50-90% of the total ^{127}I present. The mobile colloid size of $M > 0.45 \mu\text{m}$ and $0.45 \mu\text{m} > M > 10$ kDa had the highest $^{129}\text{I}/^{127}\text{I}$ ratio as for pond water and riverine water, respectively. These consequences suggest that mobile colloids greatly contribute to the transport of ^{129}I in the subsurface environment.

2A04 Development of ultrasensitive detection techniques for long-lived radionuclides with the 6 MV tandem accelerator mass spectrometry system

SASA, K.¹, TAKAHASHI, T.¹, HOSOYA, S.¹, TAKANO, K.¹, OCHIAI, Y.¹, HONDA, M.¹, OHTA, Y.¹, MATSUO, K.¹, MATSUNAKA, T.², SAKAGUCHI, A.¹, SUEKI, K.¹.

(¹Univ. of Tsukuba, ²Kanazawa Univ.)

A new horizontal-type 6 MV Pelletron tandem accelerator was

installed at the University of Tsukuba in March 2014. After 2 years of construction and preparation, the first test on multi-nuclide AMS has been performed in March 2016. The rare particle detection system on the 6 MV Pelletron tandem accelerator (Tsukuba 6 MV multi-nuclide AMS system) was designed and constructed for high-sensitivity detections of ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{41}Ca , and ^{129}I , and was also expected to measure other radioisotopes such as ^{90}Sr . The background of the system for ^{14}C measurements using the 4+ charge state and the terminal voltage $V_T = 5.0$ MV was reached down to $^{14}\text{C}/^{12}\text{C} = 2.4 \times 10^{-16}$. In addition, we successfully detected ^{10}Be , ^{26}Al , ^{36}Cl , ^{41}Ca , and ^{129}I by the multi-nuclide AMS system in 2016. In the case of ^{10}Be AMS, $^{10}\text{BeO}^-$ was extracted from the MC-SNICS and then accelerated with $V_T = 6.0$ MV. As a result, 20.3 MeV Be^{3+} was detected by a five-anode ΔE -E gas ionization detector. Isobaric interference of ^{10}B was eliminated by an absorber cell in front of the detector. The background of ^{10}Be AMS is lower than 2×10^{-15} . In this presentation, the performance of rare particle detections by the multi-nuclide AMS system will be reported.

2A05 Radiocarbon dating of the famous ancient sutras written in Asuka, Nara and Heian periods

ODA, H.¹, IKEDA, K.², YASU, H.³

(¹ISEE, Nagoya Univ., ²Faculty of Letters, Chuo Univ., ³Taga High School)

We measured radiocarbon ages of the famous ancient sutras written in Asuka, Nara and Heian periods: Myohorenge kyo attributed to Nakatomi no Kamatari (AD614-669), Gogatsu Tsuitati kyo (AD770), Uokai kyo (AD770), Hyakumanntou Darani (AD764-770), Jishinin Darani (AD764-770), Todaiji Nigatsudo Yakegyo (AD729-767), Abe no Komizumaro Gan kyo (AD871) and Chusonji Konshi Kingin Kosho kyo (AD1117-1126). The calibrated radiocarbon ages of 6 samples excluding Myohorenge kyo and Jishinin Darani were consistent with the paleographical ages. In particular, Hyakumanntou Darani was shown to be the oldest print in the world. If Myohorenge kyo truly written by Nakatomi no Kamatari, it would be the oldest existing sutra. However, the calibrated radiocarbon age was 1299-1404 [cal AD] and the theory was denied by the radiocarbon dating. Jishinin Darani had been one of the oldest existing prints. Radiocarbon dating, however, indicated that Jishinin Darani was printed in 1527-1656 [cal AD] and is not one of the oldest prints.

2A06 Search for extraterrestrial ^{60}Fe and ^{244}Pu in manganese crusts

KINOSHITA, N.¹, WALLNER, A.², FROELICH, M.B.², HOTCHKIS, M.³, FEIGE, J.⁴, PAUL, M.⁵, PAVETICH, S.², TIMS, S.G.², FIFIELD, L.K.², SCHUMANN, D.⁶, HONDA, M.⁷, MATSUZAKI, H.⁸, YAMAGATA, T.⁹

(¹Shimizu Co., ²ANU, ³ANSTO, ⁴Tech. Univ. Berlin, ⁵Hebrew Univ., ⁶PSI, ⁷Univ. Tsukuba, ⁸Univ. Tokyo, ⁹Nihon Univ.)

Nuclei of ^{60}Fe and ^{244}Pu , etc. are synthesized by multiple neutron capture reaction, r-process, in explosive environment such as supernova and neutron star merger. These nuclei present in the primordial solar system have extinguished now due to their half-lives. Our group has already found twice inputs of ^{60}Fe over a

time period of the last 10 Myr from samples collected from deep sea. The ^{60}Fe and ^{244}Pu detected on the present earth have information for r-process nucleosynthesis as well as supernova activity at vicinity of the earth. We attempted to measure time variation of ^{60}Fe and ^{244}Pu from some manganese crust samples. Fe and Be were chemically isolated from small portion of crusts. ^{60}Fe and ^{10}Be were measured using an AMS at ANU and MALT, respectively. Growth rate of crusts were determined by ^{10}Be depth profile. $^{239,240,244}\text{Pu}$ isolated from rest of the sample was measured using an AMS at ANSTO. Twice input of ^{60}Fe peak was found. In addition, more than ten counts of ^{244}Pu were detected although only one count was detected in the previous work. We will show flux and source for the nuclides using $^{60}\text{Fe}/^{244}\text{Pu}$ ratio.

2B01 Chemical reaction and property transformation of cesium during concentration reduction process using chlorination-volatilization treatment

KINOSHITA, N.¹, JIAO, F.², NINOMIYA, Y.², HONDA, M.³, SUEKI, K.³, ASADA, M.¹, KAWAGUCHI, M.¹
(¹Shimizu Co., ²Chubu Univ., ³Univ. Tsukuba)

We have investigated chemical process during concentration reduction process using chlorination-volatilization treatment. Ash samples mixed with additive of CaCl_2 , MgCl_2 , or mixture of CaO and CaCl_2 were heated at 500 - 1500 °C. After the heat treatment, XRD patterns in the slug were measured. In addition, concentration of Cs in the slug and leaching rate of Cs from the slug were determined by measuring Cs in the slug and leachate. Vaporization rate of Cs in the samples mixed with 30% of CaCl_2 reach up to around 100%. The XRD patterns show presence of CsCaCl_3 in the samples treated at 700 - 900 °C. As a result of leaching test, more than 50% of water-soluble Cs species were observed. The water-soluble Cs is assigned to be CsCaCl_3 based on the XRD patterns. The CsCaCl_3 reacts with SiO_2 and Al_2O_3 under high temperature, then CsCl produced by the reaction is vaporized. When mixture of CaO and CaCl_2 is used as an additive, complicated compound Ca-Al-Si-O-Cl is once produced. Cl produced by decomposition of the compound reacts with Cs. The CsCl is effectively vaporized because a decomposition temperature of the compound is higher.

2B02 Removal of iodide in water by bismuth-based adsorbents

LEE, S.-H. and TAKAHASHI, Y.
(Department of Earth and Planetary Science, Graduate School of Science, The University of Tokyo)

Radioactive iodine (I) is an inevitable radioactive contaminant from the processing of nuclear power plant and nuclear accident. Recent studies have shown that silver-based adsorbents such as silver oxide, silver-impregnated activated carbon and iron oxide are effective for the removal of iodine in water. Although the silver-based adsorbent has provided effective removal of iodine species in wastewater, high cost is still challenge to field application. In order to overcome this limit of silver-based material in wastewater treatment system, various adsorbents such as copper oxide, graphene oxide and manganese oxide have been investigated in an attempt to develop cost-effective adsorbents. In this study, bismuth(Bi)-based materials were studied as promising

adsorbents for the replacement of silver-based adsorbents. In order to investigate the feasibility of iodine adsorption in water, several types of bismuth-based materials such as bismuth oxide, bismuth nitrate, bismuth carbonate and bismuth-containing layered double hydroxide were prepared by facile co-precipitation method. In this talk, the adsorption feasibility of iodide which is the dominant iodine species in water by bismuth-based adsorbents will be mainly discussed with regard to adsorption efficiency.

2B03 Removal and Recovery of Cesium by Heteropolyacid Salts from High-Level Radioactive Waste

SUZUKI T., IKEZU S., SAKATE Y., KATANO T., TACHIBANA Y.

(Nagaoka Univ. of Technology)

We have proposed the novel reprocessing process with nuclide separation based on the chromatographic techniques in HCl solution system. In this reprocessing system, the high radiation nuclides are desired to be removed before sending the spent fuel solution to main process. Radioactive cesium is one of the main radiation generators in the spent fuel. It is well-known that the heteropolyacid salts have a high cesium selectivity and a high cesium adsorption capacity. We tested the ammonium molybdophosphate (AMP) and the ammonium tungstophosphate (AWP) for the cesium removal from high-level radioactive waste. The distribution coefficients in HCl solution are evaluated, and it was confirmed that both AMP and AWP have a high cesium selectivity. In addition, we synthesized the silica supported type AMP (AMP-silica) and AWP (AWP-silica) for use of column separation method. We carried out the column separation experiments (breakthrough chromatography and elution chromatography) using the simulated high-level radioactive waste (HLW) or the simulated HCl type HLW. As results, we confirmed that the cesium can be removed from HLW in the both cases (HNO_3 and HCl).

2B04 The effect of quench and its reduction for direct measurement of tritium in green tea

TOGARI, A.¹, AZUMA, K.¹, ZHOU, Q.², HATANNO, Y.³, HARA, M.³, CHIKADA, T.¹, OYA, Y.¹

(¹Grad. School of Sci., Shizuoka Univ, ²Fac. of Sci., Shizuoka Univ., ³HRC., Univ. of Toyama.)

For measuring tritium radioactivity in liquid sample, Liquid Scintillation Counter (LSC) is generally used. As an organic scintillator, 2,5-diphenyloxazole is widely applied. However, when LSC measurement is applied to green tea, a band around 350 nm – 450 nm in emission spectrum of PPO overlaps with a band around 300 – 500 nm in absorption spectra of chlorophylls and it cause the considerable reduction of counting efficiency by quenching effects. To avoid this reduction, isolating tritium water from the green tea by distillation method is typically performed. If green tea samples can be decolorized by some chemical reactions, simple and direct measurement by using LSC is possible. In this study, tritium-doped green tea samples were prepared. 30 wt% hydrogen peroxide solutions were added to each sample at different proportions. To elucidate the effect of quench and its prevention, absorption spectrophotometry was performed in addition to LSC

measurement. As a result, the reduction of quench effect was observed for samples which hydrogen peroxide was added. The counting efficiency corresponded to absorption spectra of samples around 300 nm. In addition, heating the samples also resulted in decrease of absorbance and increase of counting efficiency.

2B05 Evaluation of calibration curve in neutron activation analysis

MIURA, T.¹
(¹NMIJ)

Instrumental neutron activation analysis with the internal standardization was applied to the precise determination of Br in polypropylene resin of candidate certified reference material. The known amount of ¹⁹⁷Au was used as an internal standard to compensate for neutron flux inhomogeneity, to improve the γ ray measurement uncertainty and the linearity of the calibration curves. The neutron activations were performed using JRR3 Pn1, KUR Pn3 and KUR TcPn. In this study, the linearity of calibration curves were examined on the three neutron irradiation conditions. Unfortunately, the decrease of linearity in calibration curve was occurred in the irradiation at JRR3 Pn1. The cause of the decrease of linearity in calibration could be predicted to loss of Br by neutron irradiation. On the other hand, the analytical result of Br in the sample on irradiation at KUR Pn3 and KUR TcPn was consistent with that obtained by ID-ICPMS. The relative expanded uncertainty ($k = 2$) was 1.5 %, and it was equivalent to that of IC-ICPMS.

2B06 Radiochemical measurement of photonuclear reaction yields for photon activation analysis

OURA, Y., TERASAKI, H., YAMAGUCHI, Y., IKEDA, H., KUROIWA, N., ENDO, I., MIURA, Y., TANAKA, Y.
(Tokyo Metropolitan Univ.)

Relative reaction yields of (γ , n) reaction induced by bremsstrahlung of maximum end-point energy (E_0) of 30 MeV to 20 MeV were measured for the target nuclides with mass number from 12 to 204. Relative yields were systematically increased with an increase of target mass number (A_t), that is, a good linear correlation was obtained in double logarithmic plot. Yields for (γ , n) reactions producing isomers, however, were smaller than the systematic yield. The slope of the correlation between yields and A_t obtained by irradiation at $E_0 = 20$ MeV were slightly steeper for at $E_0 = 30$ MeV. With correlation obtained in this work, any yield ratio can be roughly estimated. And using our relative reaction yields elemental contents can be determined by a single comparator method without standard samples.

3A01 Electron spin resonance dosimetry by measuring CO₂ radicals in monkey teeth and deciduous teeth of Japanese children

OKA, T.^{1,2}, TAKAHASHI, A.³, KOARAI, K.², KINO, Y.², SEKINE, T.^{1,2}, SHIMIZU, Y.⁴, CHIBA, M.⁴, SUZUKI, T.⁴, OSAKA, K.⁴, SASAKI, K.⁴, URUSHIHARA, Y.⁵, SUZUKI, M.⁶, SHINODA, H.⁴, FUKUMOTO, M.⁷
(¹IEHE, Tohoku Univ., ²Grad. Sch. Sci., Tohoku Univ., ³Tohoku

Univ. Hospital, ⁴Grad. Sch. Dent., Tohoku Univ., ⁵NIRS, QST, ⁶IDRRR, Tohoku Univ., ⁷Dep. Mol. Path., Tokyo Med. Univ.)

The release of radioactive materials from the nuclear power plant accident in atmosphere results in an ionization radiation exposure to human beings and animals, the precise dosimetry is required on the point of view of the radiation protection and radiation safety. Electron spin resonance (ESR) method is one of a powerful tool that has been used in the study of dosimetry by measuring CO₂ radicals of teeth. In this work, we obtained the relationship between the intensity of CO₂ radical and the absorbed dose for monkey teeth and deciduous teeth of Japanese children. The detection limit for deciduous teeth of Japanese children was estimated to be 34 mGy (+- 16 mGy). We developed the novel enamel-isolation technique and applied it for monkey teeth, the detection limit of monkey teeth seems to be reduced.

3A02 History estimation of Sr-90 movement in environments by measurements of Sr-90 in hard tissues of cattle suffered from the Fukushima Daiichi Nuclear Power Plant accident

NISHIYAMA, J.¹, KOARAI, K.¹, KINO, Y.¹, SHIMIZU, Y.², TAKAHASHI, A.³, SUZUKI, T.^{2,5}, CHIBA, M.², OSAKA, K.^{2,5}, SASAKI, K.², URUSHIBARA, Y.⁴, FUKUDA, T.⁶, ISOGAI, E.⁷, OKA, T.^{1,8}, SEKINE, T.^{1,8}, FUKUMOTO, M.⁹, SHINODA, H.²
(¹Dept. Chem., Tohoku Univ., Japan, ²Grad. Sch. of Dent., Tohoku Univ., Japan, ³Tohoku Univ. Hospital, Tohoku Univ., Japan, ⁴NIRS, Japan, ⁵IRIDS, Japan, Tohoku Univ., Japan, ⁶Fac. of Sci. and Eng., Iwate Univ., Japan, ⁷Grad. Sch. of Agric. Sci., Tohoku Univ., Japan, ⁸IEHE, Tohoku Univ., Japan, ⁹Tokyo medical Univ., Japan)

We determined Sr-90 in hard tissues of cattle that were born after the Fukushima Daiichi nuclear power plant (FNPP) accident. Radioactivity of Sr-90 was measured with low background 2π gas-flow GM counter after chemical separation with nitric fuming acid. To remove ambiguity of radioactivity concentration due to the individual difference in cattle, we used specific activities which were ratio of activity to mass of the stable isotopes in the sample. Furthermore, to correct difference in individual contamination, ratio of the specific activity of the bone to that of the tooth was introduced for discussion. Almost the specific activity ratios were less than one and suggesting that the specific activity in the environment would have been decreased with an increase of the elapse time from completion of the teeth formation. To estimate the decrement, we made a model of the time course change of Sr-90 concentration in hard tissues taking the growth of the cattle into account.

3A03 Transfer mechanism of Sr-90 and Cs-137 from environment to cattle teeth at the formation period of the teeth.

KOARAI, K.¹, KINO, Y.¹, NISHIYAMA, J.¹, KANEKO, H.¹, TAKAHASHI, A.¹, SUZUKI, T.¹, SHIMIZU, Y.¹, CHIBA, M.¹, OSAKA, K.¹, SASAKI, K.¹, URUSHIHARA, Y.², FUKUDA, T.³, ISOGAI, E.¹, OKA, T.¹, SEKINE, T.¹, FUKUMOTO, M.⁴ and SHINODA, H.¹

(¹Tohoku University, ²QST, ³Iwate University, ⁴Tokyo Medical University)

We have investigated transfer mechanism of Sr-90 and Cs-137 from environment to cattle teeth. Collection sites were in the ex-evacuation area of the Fukushima-Daiichi Nuclear Power Plant (FNPP) accident. Sr-90 and Cs-137 specific activities, the ratio of the radioactivity to the weight of the stable isotopes, could provide evidences of the transfer of the nuclides. Sr-90 specific activities in teeth might correspond with those in exchangeable fractions (F1: Water soluble fractions, F2: Ion exchangeable fractions) of soils. The specific activities should change depending on the diffusion of Sr-90 and concentration of its stable isotopes in the environment. Cs-137 specific activities in the teeth were the same as those in the F2 but higher than those in the F1. Cs-137 specific activities in the F2 corresponded to teeth. Cs-137 in the F2 was main component of the exchangeable fractions. We estimate the transfer mechanism of the nuclides by comparison of the specific activities among soils, the exchangeable fractions, plants and the teeth.

3A04 Transfer of cesium to cultivated mushroom and its existing form in substrates

MURANOI, T.¹, KINO, Y.¹, ITABASHI, Y.², NAKAJIMA, T.², KORIYAMA, S.², KIMURA, E.², SHIGIHARA, T.²
(¹Dept. of Chem., Tohoku Univ., ²Kinokusu Co.)

High ability to accumulate cesium in mushrooms has been widely reported after the Chernobyl accident. After the Fukushima-Daiichi Nuclear Power Plant accident, radioactive cesium contamination of fungi has rather affected cultivated mushroom industries than other plants ones. In order to reveal a chemical behavior of the cesium in substrates for cultivated mushrooms, we analyzed the substrates by extracting water soluble component under the various conditions. We determined radioactivities of cesium-137 in extract with a high purity Ge semiconductor detector. We also determined concentration of stable cesium in those samples by an ICP-MS. We found different behavior between stable and radioactive cesium in the substrates. The results suggested that the chemical form of radioactive cesium in the substrate should be different from that of stable cesium. Since the substrate was made with trees cut just after the accident, radioactive cesium from the surface of the trees has not been sufficiently diffused into the substance. Additionally, to closely investigate the chemical form of radioactive cesium in the extracts, we separated the extracts by acid treatment and paper chromatography.

3A05 Sedimentation Processes of Radiocesium at Mountain Lakes in Gunma Prefecture, Japan

MIYASAKA, S.¹, NAGAO, S.¹, OCHIAI, S.¹, SUZUKI, K.², MORI, M.³
(¹LLRL, Univ. of Kanazawa, ²Gunma Prefectural Fisheries Experiment Station, ³Univ. of Kochi.)

The massive Tohoku Earthquake and consequent tsunamis occurred on 11 March 2011 and damaged the Fukushima Daiichi Nuclear Power Plant (FDNPP) seriously. Through the FDNPP accident, a large amount of radiocesium was released to the environment. In this study, we collected sediment cores at Lake

Haruna and Lake Onuma, which are located at high altitude in Gunma Prefecture, Japan. The concentration of ¹³⁴Cs and ¹³⁷Cs was determined by germanium detectors. We also measured the concentration of ²¹⁰Pb to estimate the sedimentation rate at each sampling site. At Lake Onuma, the ¹³⁴Cs/¹³⁷Cs activity ratio at the upper layer was almost 1.0 (decay-collected to 11 March 2011), indicating that radiocesium in the sediment samples was derived from the FDNPP accident. The inventories of ¹³⁴Cs for the outlet, inlet and center of Lake Onuma were estimated to be 2.8×10^4 , 2.2×10^4 and 1.4×10^4 Bq/m², respectively. The sedimentation rate for the outlet, inlet and center of Lake Onuma was 0.091, 0.092 and 0.060 g/cm²/y, respectively. The inventories of ¹³⁴Cs and the sedimentation rates at Lake Onuma suggested that the accumulation of radiocesium at the center resulted from atmospheric deposition. In the inlet and outlet of the lake, suspended particles adsorbed radiocesium were transported from surrounding soils and accumulated to the sediment.

3A06 Study on Migration Behavior of Suspended Solids in the Kumaki River Using Carbon-Nitrogen Isotope Ratio and Radionuclides

TAHARA, R., NAGAO, S., OCHIAI, S., HASEGAWA, T., GOTOU, M.
(Kanazawa, Univ.)

The dilapidation of Satoyama is in progress in the rural area of Japan. The decline of the agriculture and forestry workers is caused by depopulation of human and reflects to the increases in the abandonment of paddy field and forest. As a result, the possibility of erosion of soil surface occur on the abandonment area. In this study, we collected suspended solid and investigated water quality in the Kumaki River, Nanao City, Isikawa Prefecture from April 2016 to January 2017. Temporal variation of the transport of suspended solid was studied using gamma ray measurement of ¹³⁷Cs, ²¹⁰Pb_{ex} and ⁷Be deposited from atmosphere, and isotopic ratio of C (¹³C/¹²C) and N (¹⁵N/¹⁴N). The concentration of suspended particles was the highest in September after heavy rain event with 25 mm/h. This result indicated that the characteristics of suspended solid at downward flow depend on the surrounding environment and rain events.

3A07 Fractionation of Zirconium-Hafnium in Ferromanganese Crusts

INAGAKI, J.¹, SAKAGUCHI, A.¹, INOUE, M.², HABA, H.³, KASHIWABARA, T.⁴, YAMASAKI, S.¹, KIKUCHI, S.⁴, KANEKO, H.⁵, KOTANI, H.¹, TAKAHASHI, Y.⁶, USUI, A.⁷, SUEKI, K.¹
(¹Univ. of Tsukuba, ²Hiroshima Univ., ³RIKEN, ⁴JAMSTEC, ⁵JAEA, ⁶Univ. of Tokyo, ⁷Kochi Univ.)

The Zr- Hf pair is known as a “geochemical twin”, and Zr/Hf concentration ratio in various systems are theoretically the same as is found in chondrite meteorites. However, significant fractionation of the two elements between seawater and ferromanganese crusts (FMCs) has been reported. In this study, we have attempted to clarify the accumulation/fractionation mechanisms of Zr and Hf in ferromanganese crusts (FMCs) by performing Zr and Hf adsorption experiments with XAFS analysis

for synthesised and natural FMCs obtained from seamounts. The observed Zr chemical states in natural FMCs resembled those for coprecipitations with ferrihydrite and δ -MnO₂. In adsorption experiments for Zr and Hf to ferrihydrite and δ -MnO₂, it was found that Hf had larger K_D values for both minerals than Zr. In these cases, the observed Hf-O bond lengths were slightly shorter than those of Zr-O in the synthesized minerals.

3A08 Heterogeneous distribution of radionuclides in bottom sediments of the cooling pond affected by the 1986 Chernobyl Nuclear Power Plant accident

UEMATSU, S.¹, KANIVETS, V. V.², YAMASAKI, S.¹, RAHMAN, I. M. M.³, KIRIEV, S.⁴, SAKAGUCHI, A.¹

(¹CRiED, Univ. of Tsukuba, ²Ukrainian Hydrometeorological Institute, ³IER, Fukushima Univ., ⁴Chernobyl ECOCENTRE)

The cooling pond (CP) at the Chernobyl Nuclear Power Plant was contaminated with a large quantity of radionuclides due to the accident in 1986. The water level of the CP has been declining since 2014, leading to changes in water quality (e.g. water pH), affecting the solid-liquid distribution of major radionuclides, ⁹⁰Sr, ¹³⁷Cs, ²⁴¹Am and Pu. Better understanding the behaviour of radionuclides is useful to estimate the radiation effects on biota in surrounding environments. The objective of this study was to investigate the inventory of radionuclides in the bottom sediments in the CP. Core samples (depth 0-55 cm) of the bottom sediments were collected from 18 different sites in the CP. The ¹³⁷Cs and ²⁴¹Am concentrations in these sediment samples were determined by a Ge detector. The inventory of ¹³⁷Cs ranged between 0.070-21.6 MBq m⁻² and that of ²⁴¹Am varied 0.055-38.3 kBq m⁻² among these sites, showing the heterogeneous distribution of radionuclides in the bottom sediments. All the Chernobyl-derived ¹³⁷Cs existed in sediments within the depth shallower than 55 cm for all these sampling sites, except for two sites with the water depth > 9 m for which the radionuclides in sediments were detected even at 55 cm depth.

3A09 Temperature effect on soil-soil solution distribution coefficient of radiocesium

TAGAMI, K.¹, UCHIDA, S.¹, KIKUCHI, H.², KOGURE, N.²
(¹QST-NIRS, ²Tokyo Nuclear Services Co.)

A long-term after deep underground disposals of high level nuclear wastes, long-lived radionuclides may reach to the biosphere through groundwater movement. To assess radiation dose from these radionuclides, mathematical models have been used and environmental transfer parameters are needed to the models. During the long-term, climate changes are expected; therefore, it is necessary to check whether such transfer parameters are sensitive to the temperature condition or not. In this study, we measured soil-soil solution distribution coefficient (K_d) of radiocesium collected under different temperature conditions, i.e., 10, 23 and 30 °C. For this study, forty agricultural field soil samples (air-dried, <2 mm) were used and three subsamples were made for each sample. Ten mL of Milli-Q water was added to 1 g soil sample; after 24 h pre-shaking, a known amount of ¹³⁷Cs was added to the soil-water mixture and the mixture was shaken for additional seven days. The water phase (<0.45µm) was separated

and measured the ¹³⁷Cs activity with an NaI(Tl) scintillation counter. For twenty upland field soil samples, geometric mean K_d values at 10, 23 and 30 °C were 2600, 4080, 1850 L/kg, respectively; K_{as} at 30 °C were significantly lower than those at 23 °C.

3A10 Transport processes of earth surface materials based on fallout radionuclides in reservoir-catchment system

OCHIAI, S.¹, MIYATA, Y.¹, NAGAO, S.¹, MATSUKI, A.¹, UTSUNOMIYA, D.^{1,2}

(¹Kanazawa University, ²Suzu City)

To investigate the transport processes of earth surface materials in reservoir-catchment system, temporal changes in deposition of fallout radionuclides (²¹⁰Pb and ⁷Be) and their sedimentation in the irrigation reservoir (Shin-ike) were observed in Suzu City. Sedimentation fluxes of ²¹⁰Pb and ⁷Be were observed during one month with sediment trap installed in the reservoir during June 2016 to April 2017. Atmospheric deposition rate of ²¹⁰Pb and ⁷Be were also observed with a collecting pan for same intervals of sediment trap.

Deposition rates of ²¹⁰Pb and ⁷Be ranged 0.5-5.5 Bq m⁻² day⁻¹ and 3.4-25 Bq m⁻² day⁻¹, respectively, and showed clear seasonal fluctuation with high deposition rate during winter and low during summer. Sedimentation flux of ²¹⁰Pb to the reservoir bottom was much larger than ²¹⁰Pb deposition rate, suggesting that discharged ²¹⁰Pb from the catchment was major source to reservoir sediment. On the other hand, sedimentation flux of ⁷Be was almost same level with deposition rate, suggesting that the contribution of the catchment-derived ⁷Be to sediment is negligible. These differences in contribution to reservoir sediment may be associated with the timescale of material discharge from the catchment and half-lives of these nuclides.

3B01 Thermodynamic study on the Pu(IV) solubility in the presence of carbonate

KOBAYASHI, T.¹, SASAKI, T.¹

(¹Kyoto University.)

The solubility of amorphous plutonium hydroxide (Pu(OH)₄(am)) was investigated in carbonate solutions containing dilute to concentrated sodium nitrate (NaNO₃). The sample solutions were prepared by undersaturation method in the ranges of pH = 8 – 12, total carbonate concentration ($[C]_{tot}$) = 0.01 – 0.16 M, and ionic strength (I) = 0.1 – 5.0 M NaNO₃. After the given aging period at 25 °C under At atmosphere, supernatants of the sample solutions were ultra-filtrated through 10 kDa (ca. 3 nm pore size) membranes. The Pu concentration in the filtrates was determined by alpha spectrometry. The observed dependence of Pu solubility on the hydrogen ion concentration (pH_c) and $[C]_{tot}$ suggested the formation of Pu(CO₃)₄⁴⁻, Pu(CO₃)₅⁶⁻, and Pu(OH)₂(CO₃)₂²⁻ as the dominant species in neutral to weakly alkaline pH regions. The solubility at certain pH_c values and carbonate concentrations slightly increased with an increase of ionic strength. By applying the specific ion interaction theory (SIT), the solubility data at different pH_c, $[C]_{tot}$, and I were analyzed to determine the formation constants of the Pu carbonate complexes and their ion interaction coefficients. The obtained values well reproduced the

solubility data and the determined parameters are discussed in comparison with those of tetravalent actinide carbonates.

3B02 Study of the reactivity of electrodeposited U(IV)

OUCHI, K.¹, OTOBE, H.¹, KITATSUJI, Y.¹

(¹ Japan Atomic Energy Agency)

In this study, we investigated 1) the change on standing of deposition amount and 2) the catalysis by the deference of deposit stases to obtain knowledge of the reactivity of electrodeposited U(IV). 1) The change on standing of deposition amount was measured. After electrolysis was stopped, the U deposits on the electrode surface decreased regardless of the presence or absence of U(VI) ion. When the oxidation potential (+0.2 V) was applied, the decrease of the deposits was faster than that of stopping electrolysis. On the other hand, when the reduction potential (-0.35 V) was applied, the decrease of the deposits was not observed. These results indicate that the decrease of U deposits is caused by oxidative dissolution ($U(OH)_4 \rightarrow UO_2^{2+} + 2H_2O + 2e^-$). 2) U deposits of just deposition and 40 min later were formed and the rates of the reduction and deposition of U were measured. The rates of the reduction and deposition on U deposits of 40 min later were higher than that on deposits of just deposition. Thus, we found that U deposits of 40 min later have better catalytic activity.

3B03 Chemical effect on muon capture process for iron compounds

NANBU, A.¹, NINOMIYA, K.¹, INAGAKI, M.¹, KUDO, T.², TERADA, K.¹, SATO, A.¹, TOMONO, D.³, KAWASHIMA, Y.³, SHINOHARA, A.¹,

(¹Graduate School of Science, Osaka Univ., ²School of Science, Osaka Univ., ³RCNP, Osaka Univ.)

A muon can make an atomic muon orbit around a nucleus when the muon stopped in a substance. Such an atom is called muonic atom. In muonic atom formation process, muon capture probability in each atom and initial quantum state of the captured muon are strongly influenced by the chemical environment of the muon capturing atom (chemical effect). There are many studies to investigate chemical effect on muon capture process, however, the detail processes have still been puzzled. In this study, we performed systematic study on muon capture processes for iron compounds to investigate chemical effect in muon capture. The muon irradiation experiments were carried out at MuSIC-M1 beam line in RCNP, Osaka Univ. We prepared Fe, Fe₂O₃, FeS, FeO and FeCl₂ for muon irradiation samples, and measured muonic X-rays by HPGe detectors. From muonic X-ray spectra, we determined muon capture ratios and muonic X-ray intensity patterns. Per atom muon capture ratio and initial state of captured muon in each sample will be discussed on the presentation.

3B04 Production and characterization of Pm metallofullerenes using ¹⁴³Pm produced by photon nuclear reaction.

AKIYAMA, K.^{1,3}, MIYAUCHI, S.¹, AMEKURA, K.¹, ITO, K.¹, KIKUNAGA, H.², HABA, H.³, KUBUKI, S.¹

(¹Tokyo Metropolitan Univ., ²ELPH, Tohoku Univ., ³RIKEN

Nishina Center)

Promethium (Pm) metallofullerene is known as the missing lanthanide metallofullerene because all isotope of Pm is unstable. Recently, we have tried to produce Pm metallofullerenes and found that the most dominant metallofullerene is Pm@C₈₂ encapsulating trivalent Pm from the HPLC analysis. However, to obtain further information about the chemical properties of Pm metallofullerenes, it is required to employ the Pm with higher specific radioactivity because the restriction in the porosity of carbon rod used for the metallofullerene generation. Here, we report successful production of Pm metallofullerene with higher specific radioactivity by using chemically separated ¹⁴³Pm and also report the detailed HPLC elution behavior of them.

As the results of detailed HPLC analysis, it is found small differences in the HPLC retention time between ¹⁴³Pm@C₈₂ and ¹³⁹Ce@C₈₂ used as the internal standard. It is known that the HPLC retention time of lanthanide M@C₈₂ is systematically increased with increase of the atomic number for the encapsulated metal atom M. It is found that the HPLC retention time of Pm@C₈₂ is correspond to this systematics.

3B05 ⁵⁷Fe Mössbauer spectroscopic study of oxidation states of polynuclear iron hydride clusters supported by phosphines

SAKAI, Y.¹, ARAAKE, R.², TADA, M.², OHKI, Y.²

(¹Daido Univ., ²Nagoya Univ.)

Polynuclear iron hydride clusters, containing multiple iron and hydrogen atoms, are of relevance to intermediates in the biological reductive fixation of N₂, where the active state of FeMo-cofactor of enzyme nitrogenase is proposed to possess Fe-H-Fe hydrides. In the Habor-Bosch process operated in the industrial production of ammonia, metallic iron is treated with a mixture of N₂ and H₂ and the Fe-H species need to be formed prior to the N-H bond formation.

We have synthesized and characterized several novel iron hydride clusters, and then have examined the catalytic abilities for the reduction of nitrogen molecules. One of the chemical characterization tools is ⁵⁷Fe Mössbauer spectroscopy, by which the oxidation states of iron atoms of the iron hydride clusters can be elucidated non-destructively. In the present JNRS annual meeting, Mössbauer spectra are reported for the novel clusters of Fe₄(μ-H)₄(μ₃-H)₂{N(SiMe₃)₂}(PMe₂Ph)₄, Fe₆(μ-H)₁₀(μ₃-H)₂(PMe₃)₁₀ and so on, the isomer shifts of which are discussed in order to obtain clues for clarifying the catalytic roles of each iron atom sites of the clusters.

3B06 Investigation of intracellular iron in *Escherichia coli* and *Amphibacillus xylanus* lacking a respiratory chain by *in vivo* Mössbauer spectroscopy

ARIMA, H.¹, OHNO, Y.¹, KAWASAKI, S.¹, KIMATA, S.¹, TAKEDA, K.², NIIMURA, Y.¹, NAKAMOTO, T.³, TAKAYAMA, T.⁴, and SAKAI, Y.⁴

(¹Dep. of Bioscience, Tokyo, Univ. of Agriculture, ²Teachar education course, Tokyo, Univ. of Agriculture, ³Toray Research Center, ⁴Dep. of Chemistry, Daido, Univ.)

Iron must be reduced from Fe³⁺ to Fe²⁺ when organisms use it in

the cells. However, Fe^{2+} can be a generator of the most toxic reactive oxygen species, hydroxyl radical ($\bullet\text{OH}$). Thus, intracellular iron may behave as a double-edged sword in the presence of oxygen. To understand the relationship between oxygen and iron, the intracellular ratios of $\text{Fe}^{2+}/\text{Fe}^{3+}$ in two bacterial cells, *Escherichia coli* (*E. coli*) with a respiratory chain and *Amphibacillus xylanus* (*A. xylanus*) lacking a respiratory chain, which were cultured under aerobic and anaerobic conditions, were measured by Mössbauer spectroscopy (78 K). In *A. xylanus*, the both aerobic and anaerobic cells showed only IS values corresponding to Fe^{3+} , which also revealed a six-coordinate complex. In contrast, in *E. coli*, the ratio of $\text{Fe}^{2+}/\text{Fe}^{3+}$ was estimated to be 31/69 and 28/72 for the aerobic and anaerobic cells, respectively. From the IS values for Fe^{3+} , it was proved that a six-coordinate complex of Fe^{3+} was formed in the aerobic cell while a four-coordinate complex in the anaerobic cell. These results suggested that intracellular composition of Fe-chelator complexes might be altered in response to oxygen.

3B07 Dynamic Behavior of Impurity Ga in Magnetite

SUGIMOTO, T.¹, TAKENAKA, S.¹, FUJISAWA, T.¹, SATO, W.^{1,2} (¹Grad. Sch. Nat. Sci., Tech. Kanazawa Univ., ²Inst. Sci. Eng., Kanazawa Univ.)

Fe_3O_4 shows ferrimagnetism with $T_C = 858$ K, having high electric conductivity. Thus, application of this compound to spintronics has been highly expected. In our previous PAC study, we doped Fe_3O_4 with In and found that In atoms show thermal migration from the A site to the B site. This result is interesting in terms of the possibility that charge transportation can occur in coincidence with magnetic transition at T_C . Therefore, we directed our interest to Ga-doped Fe_3O_4 in expectation that more prominent dynamic behavior of the impurity atoms can be observed due to the smaller ionic radius of Ga ions than that of In ions.

0.5 at. % Ga-doped Fe_3O_4 was doped with radioactive ^{111}In ions as the PAC probe, and their hyperfine fields were investigated in a wide temperature range from 298 to 1073 K by the PAC method. Above T_C , the PAC spectrum at 873 to 1073 K ($>T_C$) shows gradual attenuation of the directional anisotropy and they were fitted with two different components: one exhibits the static perturbation by electric quadrupole interaction, and the other the dynamic perturbation showing exponential relaxation. It is considered that this relaxation reflects the relative dynamic behavior of the probe nucleus to the extranuclear fields.

3B08 Liquid-liquid extraction of element 104, Rf, in Aliquat 336/HCl system

KONDO, N.¹, KASAMATSU, Y.¹, NAGASE, M.², YASUDA, Y.¹, SHIGEKAWA, Y.¹, OUCHI, K.¹, KANDA, A.¹, NINOMIYA, H.¹, WATANABE, E.², HABA, H.³, KUBOKI, Y.³, KOMORI, Y.³, YOKOKITA, T.³, YANO, S.³, SATO, N.³, SHINOHARA, A.¹ (¹Grad. Sch. of Sci., Osaka Univ., ²Fac. of Sci., Osaka Univ., ³RIKEN Nishina center.)

The purpose of this study is to investigate the liquid-liquid extraction behavior of Rf with Aliquat 336. In our previous study, we carried out the liquid-liquid extraction experiment of Zr and Hf as a model experiment of Rf, and investigated the dependence of

distribution ratios (D) of Zr and Hf on the concentration of the Aliquat 336 by batch method. In addition, we developed the flow Injection Solvent Extraction apparatus for superheavy elements (ISE). In this work, we produced ^{261}Rf ($T_{1/2} = 68$ s) and ^{169}Hf ($T_{1/2} = 3.24$ min) in the $^{248}\text{Cm}(^{18}\text{O}, 5n)^{261}\text{Rf}$ and $^{\text{nat}}\text{Gd}(^{18}\text{O}, xn)^{169}\text{Hf}$ reactions, respectively, at the RIKEN AVF cyclotron, and carried out extraction experiment of Rf in 9.3 M HCl / Aliquat 336- CHCl_3 system. In this experiment, the reaction products were transported by the He/KCl gas-jet system to a chemical laboratory, and were dissolved in aqueous solution with the dissolution apparatus. The aqueous and organic solutions were injected to the ISE. After that, we collected two phases separately on different Ta plates and performed α -particle measurement for the samples. As a result, we obtained 63 α counts of ^{261}Rf and ^{257}No and acquired the D values of Rf.

3B09 The reversed-phase chromatography for element 105, Db in hydrofluoric acid system with Aliquat 336 resin

SATO, D.¹, MURAKAMI, M.¹, GOTO, S.¹, OOE, K.¹, MOTOYAMA, R.¹, SHIRAI, K.¹, YAMADA, R.¹, TSUCHIYA, S.⁵, MORIYAMA, T.¹, HABA, H.², KOMORI, Y.², YANO, S.², TOYOSHIMA, A.³, MITSUKAI, A.³, KIKUNAGA, H.⁴, KUDO, H.⁵.

(¹Grad. Sch. Sci. and Tech., Niigata Univ., ²Nishina Center, RIKEN, ³JAEA, ⁴Res. Center for Elec. Photon Sci., Tohoku Univ., ⁵Fac. Sci., Niigata Univ.)

In order to clarify chemical properties of Db, we studied liquid-liquid and solid-liquid extraction of Nb and Ta from HF solutions with Aliquat 336 in batch experiment. In addition, on-line chromatography experiments for Nb and Ta using ARCA were also carried out, and the extraction behaviors of Nb and Ta in 2.7 M HF and 27 M HF reproduced the results of batch experiments. On the basis of these experimental results of homologues, in this work, chemical experiment of Db was performed with 2.7 M and 27 M HF. The isotope ^{262}Db was produced in the $^{248}\text{Cm}(^{19}\text{F}, 5n)^{262}\text{Db}$ reaction with RIKEN AVF cyclotron. The reaction products were transported with He/KCl gas-jet system to laboratory and deposited on collection part in ARCA. After dissolved in 2.7 M or 27 M HF, those were loaded into micro-column filled with 52 wt% Aliquat 336 resin at a flow rate of 1.0 mL/min. Remaining species in column were stripped with 6 M $\text{HNO}_3/0.015$ M HF at a flow rate of 1.0 mL/min. These effluents were collected on different Ta disks as 1st and 2nd fraction, respectively, and were evaporated to dryness with hot He gas and a halogen lamp. Both Ta disks were picked up by robot arm, and α -activity was measured by RIDER. In 2.7 M HF, observed α -events of ^{262}Db - ^{258}Lr in 1st and 2nd fraction were 11 and 4, respectively.

3B10 An overview of the most recent gas-phase experiments with the Superheavy element Copernicium.

CHIERAN, M.^{1,2,3}, EICHLERR, I.², KRAUSB, I.², MARTZS, I.², TÜRLER, A.^{1,2}, WITTEWERY, I.², DRESSLERR, I.², PIGUETD, I.², VÖGLEA, I.², AKSENOVN, V.⁴, BOZHIKOV, G. A.⁴, CHEPIGIN, V. I.⁴, DMITRIEV, S. N.⁴, MADUMAROV, S. I.⁴, MALYSHEV, V. N.⁴, POPOV, A. A.⁴, SABEL'NIKOVA, V. A.⁴, STEINEGGER, P. I.⁴, SVIRIKHINA, I. I.⁴, VOSTOKING, K. K.⁴,

YEREMINA, V.⁴

(¹Univ. Bern, ²PSI, ³JAEA, ⁴Flerov Lab. Nucl. React.)

The study of the chemical properties of Superheavy elements (SHEs) -i.e., elements with $Z \geq 104$ -represent an extreme challenge for nuclear chemists. In fact, since SHEs experiments can be performed only at the “one-atom-at-a-time” level, they require the development of the most advanced and sophisticated setups. Here, an overview of the latest gas-chromatographic experiments with copernicium ($Z = 112$) will be presented.

P01 Classification of radiocesium absorbability by wild mushrooms using global fallout ¹³⁷Cs

TAGAMI, K.¹, UCHIDA, S.¹

(¹QST-NIRS)

Radiocesium (¹³⁴⁺¹³⁷Cs) concentrations in wild mushrooms are relatively higher than those in other wild foods according to the food monitoring results by the Ministry of Health, Labour and Welfare. However, different radiocesium concentrations were found among mushroom species in a certain concentration level area because their different habitats and growing methods, i.e. saprobic or mycorrhizal fungi. However, radiocesium concentration data in mushrooms obtained after the TEPCO's Fukushima Daiichi Nuclear Power Plant (FDNPP) accident were mainly affected by the deposition levels and number of collected mushroom species in a same deposition level areas were limited, which makes difficult to identify which mushroom species were high in concentration or not. In this study, we focused on the global fallout ¹³⁷Cs because the ¹³⁷Cs deposition amounts in Japanese soils did not differ much compared to the FDNPP origin ¹³⁷Cs. Using open available data sources, we calculated global fallout ¹³⁷Cs fraction in wild mushrooms and the values were divided by the total deposition level in each prefecture before the FDNPP accident to have aggregated transfer factor (T_{ag}). For 23 mushroom species, we could obtain more than three T_{ag} values per each mushroom species and geometric mean values of T_{ag} ranged from 1.1×10^{-3} - 6.1×10^{-2} m²/kg-fresh.

P02 Extraction and thin-layer chromatography behavior of ²¹¹At obtained by dry distillation

IKEDA, T.¹, TOYOSHIMA, A.^{2,3}, KANDA, A.¹, Zhang, Z.^{1,2}, ICHIMURA, S.⁴, KONDO, N.¹, KASAMATSU, Y.^{1,2}, YOSHIMURA, T.^{2,5}, SHINOHARA, A.^{1,2}

(¹Graduate School of Sci., Osaka Univ., ²Project Res. Center for Fundamental Sci., Grad. School of Sci., Osaka Univ., ³Advanced Sci. Res. Center, Japan Atomic Energy Agency, ⁴Fac. of Sci., Osaka Univ., ⁵Radioisotope Res. Center, Osaka Univ.)

Astatine-211 (²¹¹At) attracts a lot of attention as an alpha-emitting radionuclide suitable for targeted alpha therapy. In purification of ²¹¹At after nuclear synthesis with a solid Bi target, a dry distillation method is often utilized. However, little is known about chemical species of ²¹¹At in finally obtained aqueous solution after the distillation. Thus, in the present study, we studied extraction and thin-layer chromatography (TLC) behavior of ²¹¹At, which was obtained by the dry distillation, with and without oxidizing and reducing agents. After the irradiation of α beam on a

²⁰⁹Bi target, ²¹¹At was purified by the dry distillation. In the extraction experiment, after shaking for 2 min – 2 h, 350 μ L of both the phases (hexane and 1 M HClO₄) were precisely taken and the distribution ratio D was calculated. The TLC analysis was also performed for the ²¹¹At samples in distilled water. As a result, the D values of ²¹¹At into hexane increased with increasing shaking time under the conditions without oxidizing and reducing agents. On the other hand, ²¹¹At was not extracted into hexane under the oxidative and reductive conditions. In the TLC experiments, different 2 peaks of ²¹¹At with around 0 and 0.9 of R_f values were observed. These results suggest that neutral species with low polarity and ionic one with high polarity exist in distilled water.

P03 Development of metallofullerene separation by chemical reduction-2

TANAKA, K.¹, AKIYAMA, K.¹, HABA, H.², KUBUKI, S.²

(¹Tokyo Metropolitan Univ., ²RIKEN, Nishina center.)

Endohedral metallofullerene (EMF) is expected to be applied for radiopharmaceuticals due to their encapsulation character. For such application, radioactive EMF is necessary to have high specific radioactivity. It is well known that the large amount of empty fullerenes, such as C₆₀ and C₇₀, are produced as a by-product during the EMF production. To obtain the radioactive EMFs with high specific radioactivity, it is required to develop the efficient separation method for trace amount of EMFs from by-products instead of the conventional HPLC separation method. Recently, we developed a new separation method combining the reduction of EMFs using an electron donor and the difference of solubility in a polar solvent between reduced EMFs and empty fullerenes. In this session, we will report the quantitative results of this separation method using radiotracer technique.

Radiotracer of Ce-139 was employed for the EMF production. Produced Ce-139 EMFs were dissolved to CS₂ and then added triethylamine (TEA). After that, acetonitrile (MeCN), methanol (MeOH), and mixed solution of MeCN/MeOH were used for the separation of reduced EMFs from empty fullerenes. Gamma-ray measurement was performed for each separated sample. HPLC analysis was also performed to investigate recovery rate of each EMF and empty fullerene species. As the results of γ -ray measurement and HPLC analysis, the best recovery rate was found in the case of MeCN/MeOH

P04 Developments of time of flight prompt gamma-ray analysis and its application

TOH, Y.¹, HUANG, M.^{1,2}, EBIHARA, M.³, SEGAWA, M.¹, TUCHIYA, H.¹, MAEDA, M.¹, KIMURA, A.¹, NAKAMURA, S.¹

(¹JAEA, ²IMP, ³TMU.)

The Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) has been developed for measurements of prompt gamma-ray analysis (PGA), nuclear cross-section data and nuclear astrophysics at BL04 of the Material and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). Two cluster-Ge detectors and eight coaxial-Ge detectors with BGO Compton-suppression shields was installed at the flight length of 21.5m in ANNRI. The

detector-array is designed to provide high gamma ray energy resolution and high detection efficiency. We have developed a time-of-flight prompt gamma-ray analysis (TOF-PGA) by using an intense pulsed neutron beam. We have conducted several experiments of standard samples to establish the dead-time correction method of TOF-PGA. The correction method was verified using experimental measurements of Ni-based super alloys. Presentation will describe the detail of the experiments and new device (neutron filter), the current status of the developments and future prospects.

P05 The determination of uranium ratios in individual environmental particles

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

In the previous study, we developed a combination method of particle manipulation and secondary ion mass spectrometry (SIMS) for removing the interferences from other elements in other particles. Since the particles were selected randomly and manipulated, it caused the problem not to cover the range of $^{235}\text{U}/^{238}\text{U}$ isotope ratio in the sample. In this study, we improved the procedure of analyzing uranium isotope ratios in individual particles by SIMS to cover the range of $^{235}\text{U}/^{238}\text{U}$ in the sample.

70 uranium particles in an environmental sample were manipulated and screened by using an automated particle measurement (APM) software of the SIMS instrument. After the APM measurement, several uranium particles were selected to cover the range of $^{235}\text{U}/^{238}\text{U}$ and accurate uranium isotope ratios were determined by SIMS with microprobe mode, that mode is capable to measure more accurate isotope ratios compared with APM measurement.

APM measurement resulted the range of $^{235}\text{U}/^{238}\text{U}$ from 0.001 to 0.053. This distribution of isotope ratio is useful information for selecting particles to be measured by SIMS with microprobe mode. APM measurement performed simplified measurement to know the range of isotope ratios in the samples. In this improved procedure, we could cover the range of $^{235}\text{U}/^{238}\text{U}$ in the sample even if approximately 10 uranium particles were measured with microprobe mode

P06 The Environmental Impact of the Fukushima Dai-ichi Nuclear Power Plant Accident on Land of Miyagi Prefecture

ISHIKAWA, Y.¹, TAKAMURE, T.¹, HATAKEYAMA, N.¹, ARAI, Y.², TAKAHASHI, M.¹, ANDO, T.¹
(¹Miyagi Pref. Environ. Rad. Monitoring Center, ²Miyagi Pref. Nucl. Safety Division)

Miyagi Prefecture, adjacent to the north of Fukushima Prefecture, has been contaminated with radioactive materials transported by the plumes from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, occurred on March, 2011. The Environmental Impact of the accident on Land of Miyagi Prefecture was investigated. Radioactivities of ^{137}Cs and other radionuclides in airborne dusts, rain and dry fallout, surface soils, plants as bio-indicators, and other vegetation were measured by gamma-spectrometry using Ge-detectors. By measurement of fallout and soils, monthly depositions and inventories of ^{137}Cs

were determined, and then compared with radioactivities of plants (wormwood leaves). ^{137}Cs deposition increased to about 10,000 Bq/m² after the accident, from about 0.1 Bq/m² before the event. As a result, ^{137}Cs inventory in soil after the accident increased by two orders of magnitude from 100-200 Bq/m² before the accident. Similarly, ^{137}Cs concentrations in plants increased by two orders of magnitude from 0.1-1 Bq/kg (wet) before the accident. Correlation among radioactivities of ^{137}Cs in evaporation residues of fallout samples, weight of those and radioactivities of ^{137}Cs in air were also investigated.

P07 Study on a Determination Method of Radioactive Antimony in Soil near Fukushima Daiichi Nuclear Power Plant

MIYAZAWA, N.¹, UESUGI, M.², YOKOYAMA, A.³
(¹Graduate School of Natural Science and Technology, Kanazawa University, ²Advanced Science Research Center, Kanazawa University, ³Institute Science and Engineering, Kanazawa University)

A large amount of radionuclides was released into the atmosphere due to the accident at Fukushima Daiichi Nuclear Power Plant (FDNPP) in 2011 and deposited on the soil. One of the radionuclides is ^{125}Sb ($T_{1/2} = 2.75$ y). The deposited amount of ^{125}Sb and its distribution are possible to be useful in estimating production of the nuclide, unknown phenomena such as its chemical reactions, and its release to the environment. The emission amount of ^{125}Sb is relatively small and counting photon in peak area is disturbed by γ - rays from abundant radioactive cesium. The quantitative analyses are seldom reported. Therefore, in this study, we established a quantitative method of ^{125}Sb by removing cesium. We studied two methods, one of which utilizes removing Cs with ammonium phosphomolybdate adsorption and concentrating Sb by iron co-precipitation, and the other does removing Cs and concentrating Sb with an anion exchange resin. The chemical recovery was examined by addition of ^{124}Sb ($T_{1/2} = 60.3$ y) tracer. Using the examined method, we succeeded in quantifying the activity of ^{125}Sb in contaminated soil near FDNPP.

P08 Rice cultivation test for decontamination of the paddy soil contaminated with radioactive cesium

TANIGUCHI, D.¹, DEZAWA, Y.¹, YANAGA, M.², GOTO, S.³, SERA, K.⁴
(¹Grad. of sci., Shizuoka Univ., ²Fac. of Sci., Shizuoka Univ., ³Takizawa Lab., JRIA., ⁴Cyclo. Res. Center, Iwate Medical Univ.)

The soil of many fields was contaminated with radioactive cesium due to the accident at the Fukushima Daiichi Nuclear Power Plant. In the present work, cultivation tests with soil artificially contaminated with ^{137}Cs were performed. Fifteen pots which the contaminated soil was put in were divided into five groups. Stable isotope of Cs was added to two groups before cultivation or midsummer drainage. The radioactivity of harvested rice plant (brown rice, leaf and stem), was measured with a Ge semiconductor detector. The highest value of radioactivity of Cs was found in the group of pots in which stable isotope of Cs was added to the soil before cultivation. The second value was found in

the group to which Cs carrier was added before midsummer drainage. This means that the earlier stable isotope of Cs was added, the higher radioactivity of brown rice was transferred to rice plant from soil. It is also considered that the added stable Cs ions were a competitive relationship with radioactive ions.

P09 High sensitivity analysis of airborne particulates

SHIKIMI, J., NITTA, W., AKIYAMA, M., SUZUKI, K., ISOGAI, K.
(Japan Chemical Analysis Center)

In recent environmental radioactivity monitoring, ^{134}Cs and ^{137}Cs in airborne particulates are often undetectable because of their low concentrations, which unables quantitative evaluation. In this study, an ultra high volume air sampler was used to increase the sample volume and lower the detection limits for airborne particulates. Airborne particulates were collected by using an ultra high volume air sampler JL-900 manufactured by SENYA (hereafter referred to as "SNOW WHITE") in Chiba, Japan. About 60,000 m³ of air was filtered with a flow rate of 680 m³/h during each sampling period of about 3.5 days. The airborne particulates was collected on a filter paper and packed into a plastic container for gamma-ray measurement by Ge semiconductor detector. This study reported about airborne particulates which were collected and analyzed from February 2016 to February 2017. The obtained radioactivity concentrations of ^{134}Cs and ^{137}Cs were ND ~ 0.0057 mBq/m³ and 0.0022 ~ 0.040 mBq/m³, respectively. Since SNOW WHITE has about 20 times higher sampling capacity compared with a conventional low-volume air sampler, ^{134}Cs and ^{137}Cs were detected in most of samples collected even for short sampling period of 3.5 days. This also enables us to evaluate the short term trends of radioactivity concentrations in air. However ^{134}Cs were still undetected in several samples, it will be necessary to take improvements such as increase sampling volume.

P10 Air dose rate monitoring at the portable radiation monitoring stations in Fukushima Prefecture

TANAKA, H., MIYATA, S., SATO, S., NITTA, W., ISOGAI, K.
(Japan Chemical Analysis Center)

Air dose rates have been continuously monitored at the 80 portable radiation monitoring stations located around Fukushima Dai-ichi Nuclear Power Plant. Sixty of the monitoring stations were equipped with NaI scintillation detectors, and the rest with silicon detectors manufactured by Fuji electric Co., Ltd. In the monitoring results measured at the 80 monitoring stations during 2012 ~ 2016, the air dose rates decreased over time due to radioactive decay of ^{134}Cs and ^{137}Cs . This trend was conspicuous at the stations which had relatively higher deposition of radiocesium and high dose rates. Short-term changes in air dose rates possibly caused by natural radionuclides were also observed at many stations. It is well known that the rain-out and wash-out of radon progenies can cause the increase of air dose rate near ground surface. In this study however, decreases of air dose rate during rainfall were often observed at many stations. This means that the gamma-rays emitted from radiocesium deposited in soil were shielded by water layer on the ground temporarily produced by rainfall, resulting in the decrease of air dose. The contribution of the shielding was large enough to cancel out the increase of dose

caused by radon progenies. At most of the sites, air dose rates conspicuously decreased due to the decontamination activities performed around the sites.

P11 Preparation of thick ^{238}U metallic target using sputtering method and its irradiation test

KAJI, D.¹, MORIMOTO, K.¹, HABA, H.¹

(¹RIKEN, Nishina Center for Accelerator-Based Science)

We newly developed the sputtering system, which was installed magnetron source with 1-inch size, for preparing thick ^{238}U metallic target. Under the condition, such as an argon-gas flow rate of 6 sccm and a pressure of 7.3 Pa inside the chamber, we checked the sputtering rate of the metallic ^{238}U with tuning RF-power for sputtering. Then, we obtained the sputtering rate of 0.263 μg/s for 70W and 0.362 μg/s for 100W, respectively. Finally, we succeeded to prepare the thick ^{238}U metallic target with about 500 μg/cm² thickness on 3 μm Ti-backing foil. The ^{238}U target was utilized for studying on production and decay properties of Cn (Z=112) isotopes via the reaction $^{48}\text{Ca}+^{238}\text{U}\rightarrow^{286}\text{Cn}^*$ using a new gas-filled recoil ion separator GARIS-II.

In this report, we will give details about the preparation of thick ^{238}U metallic target and its irradiation test.

P12 The transfer of radiocesium from paddy soil and water to rice plants

DEZAWA, Y.¹, TANIGUCHI, D.¹, YANAGA, M.², GOTOU, S.³, SERA, K.⁴

(¹Grad. of Sci., Shizuoka Univ., ²Fac. of Sci., Shizuoka Univ., ³Takizawa Lab., JRIA.,

⁴Cyclo. Res. Center, Iwate Medical Univ.)

The Fukushima nuclear power plant accident released a large amount of radioactive material into the environment. The soil of paddy field was also contaminated with radioactive cesium. So, it is concerned that rice plants are contaminated with radiocesium although there are no reports that radioactivity in the rice exceeded the standard value for these several years. In the present work, cultivation tests were performed in order to confirm which radioactive cesium in soil or irrigation water would be transferred to rice. At first, non-contaminated soil collected in Shizuoka-city was artificially contaminated with ^{137}Cs . After pouring water and raising rice plants in Wagnel pots with the contaminated soil, ^{134}Cs was added to water. Harvested brown rice was loaded into a U8 container and the radioactivity of ^{137}Cs and ^{134}Cs in the brown rice was measured by a Ge semiconductor detector. The uptake quantity of ^{134}Cs in brown rice was three times larger than that of ^{137}Cs . It is suggested that the radiocesium in the brown rice mainly transferred from the water. It is also found that the uptake quantity of ^{134}Cs added after mid-summer drainage was higher than that of one before.

P13 Investigation on solvent extraction of astatine for ^{211}Rn - ^{211}At generator

SHIN, Y.¹, KAWASAKI, K.¹, YAMADA, N.¹, YOKOYAMA, A.¹, WASHIYAMA, K.¹, NISHINAKA, I.²

(¹Kanazawa Univ., ²QST)

^{211}At is a promising nuclide among various α -emitters for targeted α therapy. It has high α -particle energy and the emission probability of α -ray is about 100%. However, ^{211}At is produced by an accelerator and the half-life of ^{211}At is only 7.2 hour. It is too short to deliver to remote areas from accelerator facilities. In order to use astatine in wider ranges we develop a ^{211}Rn - ^{211}At generator. In the previous study, ^{211}At produced by ^{211}Rn decay in dodecane solution can be back-extracted by 90% ethanol aqueous solution with N-bromosuccinimide (NBS), while the mechanism of astatine extraction with NBS is not known. In this study we assumed that Br_2 generated by NBS is effective for astatine back-extraction and used bromine water instead of NBS. ^{211}At was produced in the $^{209}\text{Bi}(\alpha, 2n)$ reaction by AVF cyclotron at RIKEN. ^{211}At was separated from Bi target by heating and collected in dodecane. ^{211}At was back-extracted by 90% ethanol aqueous solution with Br_2 of various concentrations. After extraction, the radioactivities of ^{211}At in water phase and organic phase were measured by liquid scintillation counter to obtain its distribution ratios. The results are similar to those of NBS, but include the best astatine back-extraction ratio obtained so far.

P14 Development of cavity ring-down spectroscopy apparatus for chemical identification of carrier-free radioiodine and astatine in gas phase

TOYOSHIMA, A.¹, AOYAGI, N.²

(¹Advanced Science Research Center, Japan Atomic Energy Agency, ²Nuclear Science and Engineering Center, Japan Atomic Energy Agency)

Carrier-free radiotracers are often utilized in lots of research fields because of its utility based on high sensitivity of nuclear radiation. However, it is difficult to identify their chemical forms by standard UV/vis and IR spectroscopy because of their micro quantities. These are deduced from behavior of the radiotracers in partition methods such as chromatography and solvent extraction. In the present study, we will report our status on development of an apparatus for cavity ring-down spectroscopy applied to the measurement of carrier-free radioactive iodine and astatine in a gas phase.

P15 Observation of Zinc Vacancy-Hydrogen Complexes in ZnO by Means of Positron Annihilation Spectroscopy

SHIMIZU, H.¹, SATO, W.^{1,2}, MIHARA, M.³, FUJISAWA, T.¹, FUKUDA, M.³, MATSUTA, K.³

(¹Grad. Sch. Nat. Sci., Tech., Kanazawa Univ., ²Inst. Sci. Eng., Kanazawa Univ., ³Grad. Sch. Sci., Osaka Univ.)

Zinc Oxide (ZnO) is highly expected for applications to functional devices because of its wide band gap of 3.4 eV. ZnO shows persistent photoconductivity, which causes the degradation of performance of devices such as thin film transistors used in liquid crystal displays. Zn vacancy-hydrogen complexes ($V_{\text{Zn}} + n\text{H}$) are a possible candidate as one of the cause of the degradation. Their removal from ZnO is essential, but their thermal stability has not been fully understood yet. In this work, positron annihilation lifetime spectroscopy was employed for the observation of the formation and desorption processes of $V_{\text{Zn}} + n\text{H}$ in single crystal

ZnO in relation to heat treatment at various temperatures. In order to highlight the behavior of $V_{\text{Zn}} + n\text{H}$ complexes, the sample was implanted with $^1\text{H}^+$ using an electrostatic accelerator. After the implantation, the positron lifetime became shorter, which suggests the implanted hydrogen atoms were captured by V_{Zn} to form the complexes. At 473 K, it was suggested by an increase of positron lifetime that most of hydrogen atoms dissociate from V_{Zn} ; however, the lifetime hardly changed above 573 K. In the present session, the constant positron lifetime is discussed based on complementary studies for unimplanted single crystal ZnO by means of positron annihilation lifetime spectroscopy and Doppler-broadening spectroscopy.

P16 Electronic and chemical states of iron in ancient buried trees revealed by ^{57}Fe Mössbauer spectroscopy

YAMAUCHI, S.¹, KURIMOTO, Y.¹, SAKAI, Y.²

(¹Akita Pref. Univ., ²Daido Univ.)

The electronic and chemical states of Fe species in ancient trees (umoregi trees) excavated from the foothills of Mt. Chokai in Japan were characterized using ^{57}Fe Mössbauer spectroscopy and complementary analytical techniques. The umoregi wood samples were prepared from the stems of four tree species. Japanese cedar and Japanese zelkova umoregi woods were light brown and dark green, respectively. Japanese oak and Japanese chestnut umoregi woods exhibited pure black tone. Treatment with oxalic acid removed these characteristic color tones from the umoregi woods. Infrared photoacoustic spectra indicated that the degradation of the umoregi woods has not proceeded drastically. Mössbauer spectra of the umoregi woods were recorded at 293 K and 78 K, and their absorption line shapes were analyzed using a curve-fitting method. The resulting Mössbauer parameters confirmed that Fe was trivalent and its spin momentum number was 5/2 (i.e. high-spin state). From this comprehensive investigation, it was suggested that Fe^{3+} in Japanese oak and Japanese chestnut umoregi woods forms multinuclear complexes.

P17 Particle size distribution of atmospheric aerosols including ^7Be and major ionic species

MATSUBARA, R.¹, HIWATASHI, M.², KONDO, A.¹, and MURAMATSU, H.¹

(¹Fac. of Edu., Shinshu Univ., ²Grad. Sch. of Edu., Shinshu Univ.)

It has been desired to obtain more knowledge concerning migration, diffusion, and transport processes of airborne radioactive materials after the Fukushima nuclear accident. In order to estimate the chemical form of atmospheric aerosols as a carrier of radioactive materials, we have carried out the measurement of the particle size distribution of atmospheric aerosols attached ^7Be and determination of the components of ionic species contained in the atmospheric aerosols, by using a 12-stage low-pressure cascade impactor (Tokyo Dylec Co., LP-20) and a high performance liquid chromatography (HPLC, Shimadzu). We found that excellently dominant ionic species were SO_4^{2-} and NH_4^+ , and their mass median aerodynamic diameter (MMAD) were 0.53-0.68 μm and 0.54-0.74 μm , respectively, which are quite similar or almost identical to the activity median aerodynamic diameter (AMAD) of the ^7Be -aerosols, 0.44-0.67 μm . So, we

conclude that aerosols containing NH_4^+ and SO_4^{2-} would be the potential transport medium for ^7Be .

P18 The evaluation of adsorption properties of metal ions on Sr selective resins and application to analysis of Sr-90 in fish bone ash sample

MIURA, T.¹, MINAI, Y.²
(¹NMIJ, ²Musashi Univ.)

A simple analytical method was developed for the determination of ^{90}Sr in fish bone ash samples. The adsorption behaviors of 26 metal ions on Eichrom Sr Resin and IBC Advanced Technology AnaLig Sr-01 resin were examined. The obtained distribution coefficients show that Sr Resin has suitable selectivity. The proposed method consists of co-precipitation with calcium phosphate and purification by Sr Resin. After ingrowth, ^{90}Y was measured using a liquid scintillation counter. The proposed method was validated by radiometric analysis of ^{90}Sr in certified reference materials (NIST SRM 1400 bone ash, and IAEA-156 Radionuclides in clover). Finally, the proposed method was applied for interlaboratory comparison of ^{90}Sr analysis in fish bone ash sample. The analytical results by proposed showed acceptable z score.

P19 Quality evaluation of $^{99\text{m}}\text{TcO}_4^-$ solution from ^{99}Mo generated with accelerator induced neutrons

KAWABATA, M.^{1,2}, NAKAHARA, Y.³, HATSUKAWA², Y., MOTOISHI, S.^{1,2}, SAEKI, H.^{1,2}, OHTA, A.^{1,2}, SHIINA, T.¹, KAWAUCHI, Y.¹, HASHIMOTO, K.², AND NAGAI, Y.²
(¹Chiyoda Technol Corp., ²QST, ³FUJI FILMRI Pharma Co., Ltd.)

^{99}Mo can be produced via the $^{100}\text{Mo}(n,2n)$ reaction using accelerator generated neutrons where daughter nuclide $^{99\text{m}}\text{Tc}$ is thermo-chromatographically separated from a relatively large sample mass of MoO_3 . Radiopharmaceutical quality of the obtained $^{99\text{m}}\text{Tc}$ pertechnetate solution was assessed to ensure that the production method could be an alternative to a fission production. The result showed that the quality of the $^{99\text{m}}\text{Tc}$ pertechnetate solution met the United States Pharmacopeia requirements, hence it was compatible to commercially available $^{99\text{m}}\text{Tc}$ pertechnetate solution. The non-radioactive molybdenum content, which could have been present from the $^{100}\text{MoO}_3$ raw material, was far below than a tolerance dose given in international guidelines. Good labelling yields were obtained for the four radiopharmaceutical agents (MIBI, ECD, MAG3 and MDP). This result strongly supports that the production of ^{99}Mo obtained via the $^{100}\text{Mo}(n,2n)$ reaction using accelerator induced neutrons followed by thermo-chromatographic separation of $^{99\text{m}}\text{Tc}$ can be a promising substitute for the fission product ^{99}Mo .

P20 Influence of number of introduced molecules on isothermal chromatography of Zr and Hf chloride

SHIRAI, K.¹, GOTO, S.¹, OOE, K.¹, KUDO, H.²
(¹Grad. Sch. Sci. Tech., Niigata Univ., ²Fac. Sci., Niigata Univ.)

In a gas phase chemistry of superheavy elements, it is believed that there is an empirical linear correlation between a standard

sublimation enthalpy and an adsorption enthalpy on quartz surface for a volatile species of interest, and the sublimation enthalpy is deduced from the adsorption enthalpy obtained in single-atom experiments. Although the volatility sequence which was obtained from the results in on-line experiments was $\text{ZrCl}_4 \geq \text{RfCl}_4 > \text{HfCl}_4$, especially, the relation between ZrCl_4 and HfCl_4 differs from the relation of sublimation expected from their vapor pressure curves in the macro-scale. In this study, we performed isothermal gas chromatography with various number of molecules to clarify cause of the difference in behavior of adsorption.

ZrOCl_2 or HfOCl_2 solution of a certain concentration including radiotracers of ^{88}Zr or ^{175}Hf was once transformed into oxide, after that, the oxide was heated with CCl_4 at 600 °C and turned into a chloride. The formed chloride was collected on carbon filter at a chloride-collection part, and then was evaporated at 400 °C. The volatile species passed through an isothermal column at various temperatures, and cumulative yields were obtained with γ -ray measurement. We applied a chromatography model that the volatile species can adsorb on sites of two types, the original surface and the molecular layer of the adsorbed species. In the symposium, the difference of mechanism to pass through the column between macro- and tracer-scale will be discussed in more detail.

P21 Temporal variation of $^{36}\text{Cl}/\text{Cl}$ ratio in rainwater at Tsukuba before and after the FDNPP accident

OCHIAI, Y.¹, SASA, K.¹, TAKAHASHI, T.¹, TOSAKI, Y.², MATSUMURA, M.¹, ABE, S.¹, HOSOYA, S.¹, TAKANO, K.¹, SUEKI, K.¹
(¹Univ. of Tsukuba, ²AIST)

On March 11, 2011, a huge earthquake and the following tsunami severely damaged the Fukushima Daiichi Nuclear Power Plant (FDNPP). Various nuclear reaction products were subsequently emitted from the accident (e.g. ^{35}S). This study focuses on ^{36}Cl which is also expected to have been produced inside the reactor and released into the environment. At the University of Tsukuba, the $^{36}\text{Cl}/\text{Cl}$ ratios in monthly rainwater samples were measured with accelerator mass spectrometry (AMS). In this study, we measured the $^{36}\text{Cl}/\text{Cl}$ ratios in rainwater collected from November 2010 to December 2011, by using the AMS system at the University of Tsukuba. As a result, it was suggested that the ratio in March 2011 was higher than in February 2011. In the poster, we will report on detailed analysis and the comparison with the seasonal trend of $^{36}\text{Cl}/\text{Cl}$ ratios observed for 2004-2009 and the variation of $^{129}\text{I}/^{127}\text{I}$ ratio during 2010-2011.

P22 Migration of radiocesium in suspended matter by rainfall in Tamagawa River watershed

OKUMURA, S.¹, SHIMONO, A.², FUJII, K.¹, OCHI, K.³, HAGIWARA, K.², KOIKE, Y.²
(¹ Grad. Sch. of Sci. and Tech., Meiji Univ., ²Sch. of Sci. and Tech., Meiji Univ., ³JAEA)

A large amount of radiocesium released into the atmosphere by the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident was deposited on urban area. The continual monitoring of radiocesium at the river watershed is important for estimating

environmental impact by migration of FDNPP-derived radiocesium through a river system. Activity concentrations of radiocesium were determined for the suspended, dissolved and sediment samples collected at the Tamagawa River watershed. Among the radiocesium derived from the FDNPP in river water, radiocesium adsorbed on suspended matter decreased in the river watershed. High concentration of suspended radiocesium has been reported by rainfall events because of increase of the amount of suspended matter. In this study, activity concentrations of ^{137}Cs in suspended matter were observed at regular intervals from the start to the end of rainfall.

P23 Measurement of Internal Fields in Co and Mn Co-doped ZnO by Means of ^{57}Co Emission Mössbauer Spectroscopy
MORIICHI, R.¹, KANO, Y.¹, SATO, W.^{1,2}
(¹Grad. Sch. Nat. Sci., Tech., Kanazawa Univ., ²Inst. Sci., Eng., Kanazawa Univ.)

We measured internal fields in 6.25 at.% Co- and 6.25 at.% Mn-co-doped ZnO (CMZO) synthesized by a solid state reaction by means of ^{57}Co emission Mössbauer spectroscopy. Two doublets were observed in the spectrum measured at room temperature: one is assignable to $^{57}\text{Fe}^{2+}$ ($\leftarrow^{57}\text{Co}$) substituting for defect-free Zn site in ZnO lattice and the other to $^{57}\text{Fe}^{3+}$ ($\leftarrow^{57}\text{Co}$) in substitutional Zn sites adjacent to a Zn vacancy. However, below 100 K, spectra cannot be fitted only with the two doublets, and we assumed the emergence of a magnetic component. The relative intensity of this component is increased with decreasing temperature. The temperature dependence for the Mössbauer parameters are similar to those obtained from the spectra for 5 at.% Co- and 5 at.% Mn-co-doped ZnO. In addition, we investigated the effect of co-doping method by magnetization measurements for CMZO, 11 at.% Co-doped ZnO (CZO), and 6 at.% Mn-doped ZnO (MZO). Unlike CZO and MZO, CMZO shows slightly different trend from that expected for paramagnetism. The results of ^{57}Co emission Mössbauer spectra and magnetization measurements imply that the present co-doping method can realize the emergence of dilute magnetism in CMZO.

P24 Synthesis of $\text{Y}@\text{C}_{60}$ by nuclear recoil
ITO, Y.¹, AKIYAMA, K.¹, KUBUKI, S.¹, HATSUKAWA, Y.²
(¹Tokyo Metropolitan Univ., ²QST.)

Endohedral metallofullerene: $\text{M}@\text{C}_{60}$ is one of the most interesting metallofullerene species because of their symmetric structure and chemical properties. The most interesting chemical properties of $\text{M}@\text{C}_{60}$ is the differences in the solubility to organic solvents from other metallofullerenes such as $\text{M}@\text{C}_{82}$. It is known that $\text{M}@\text{C}_{60}$ is soluble to aniline and pyridine but insoluble to good solvent for general metallofullerene species such as toluene and CS_2 . In previous study, we synthesized $^{85}\text{Sr}@\text{C}_{60}$ using the nuclear recoil of $^{86}\text{Sr}(n, 2n)^{85}\text{Sr}$ reaction and found that the production yield of $^{85}\text{Sr}@\text{C}_{60}$ depend upon the particle size of Sr salts used as the target. Here, we report the production of $^{88}\text{Y}@\text{C}_{60}$ using the nuclear recoil of $^{89}\text{Y}(n, 2n)^{88}\text{Y}$ reaction.

As a result, the radioactivity of ^{88}Y observed in the aniline solutions derived from $^{88}\text{Y}@\text{C}_{60}$ was approximately estimated to be 4% in the total radioactivity of each sample for all aniline

solutions examined in this work and it was found that the production yield of $^{88}\text{Y}@\text{C}_{60}$ for each sample was almost similar in spite of different particle size. From the observed SEM image, the similarity in the production yield of $^{88}\text{Y}@\text{C}_{60}$ is expected to be caused by the aggregation of Y_2O_3 nanoparticles.

P25 Study of the Electronic Structure of a Spin Crossover Iron(II) Complex with Acidic Protons
N. OKAWA, T. SHIGA, H. OSHIO
(Univ. of Tsukuba)

Complexes that can be switched by various external fields in addition to being triggered by deprotonation of the ligands can be expected to be new switching materials. In this work, our aim is to control the physical properties of our complex using protonation or deprotonation of the ligands. We have synthesized a protonated iron(II) complex $[\text{Fe}^{\text{II}}(\text{H}_2\text{L})_2](\text{BF}_4)_2$ (**1**) and a deprotonated iron(III) complex $(\text{PPh}_4)[\text{Fe}^{\text{III}}(\text{L})_2]$ (**2**) using the diprotic tridentate ligand H_2L . The electronic structures of the iron ions in both complexes have been determined by single crystal X-ray analysis, magnetic and Mössbauer measurements. Single crystal X-ray structure analysis suggested that **1** was in the Fe(II) state and the ligand was protonated due to existence of two BF_4^- anions, whereas **2** was in the Fe(III) state and the ligand was deprotonated due to existence of one PPh_4^+ cation. Magnetic susceptibility and Mössbauer measurements revealed that **1** showed spin crossover behavior with $T_{1/2 \uparrow} = 150.3$ K, in which incomplete spin crossover was observed. On the other hand, **2** was in the low-spin state in all temperature measured. We also discuss light induced excited spin state trapping (LIESST) behavior and solution properties.

P26 A study on estimation of dissolved oxygen content in Tokyo Bay using chemical states of sedimentary iron as an indicator.
KOMORI, M.^{1,2}, SHOZUGAWA, K.¹, MATSUO, M.¹
(¹Graduate School of Arts and Sciences, The University of Tokyo, ²Yokohama Environmental Science Research Institute)

Hypoxia is a big problem particularly in inner bay such as Tokyo Bay. To understand and solve the problem, it is important to know when or where hypoxia occurred. However, the data of water quality in Tokyo Bay are absolutely lacking. Therefore, we attempted to estimate the dissolved oxygen content in Tokyo Bay using chemical states of sedimentary iron as an indicator.

We collected sediment cores off the coast of Yokohama (flat seafloor) and Makuhari (flat seafloor and dredged trench) in Tokyo Bay. The samples were sliced every 2 cm and analyzed for the chemical states of iron by Mössbauer Spectroscopy.

As a result of the analysis, all samples of Yokohama and the surface layer of Makuhari tend to contain less Fe^{2+} and more ferromagnetic or antiferromagnetic iron. This indicates that seawater in Yokohama (center of Tokyo Bay) may be more oxic than that in Makuhari (inner part of Tokyo Bay), and seawater in Tokyo Bay may have been improved recently. This is consistent with monitoring data of dissolved oxygen content in seawater.

In addition, even though sampling spots are really close, sediment at dredged trench of Makuhari tends to contain more Fe^{2+} than that at flat seafloor, which indicates that dredged trench is one

of the causes of hypoxia.

P27 Evaluation for specific radioactivity of ^{67}Cu for cancer therapy, produced with accelerator neutrons by deuterons

HASHIMOTO, K.¹, KAWABATA, M.^{1,2}, SAEKI, H.^{1,2}, SATO, S.², TSUKADA, K.³, SUGO Y.¹, NAGAI Y.¹, HATSUKAWA, Y.¹, ISHIOKA, N. S.¹

(¹National Institutes for Quantum and Radiological Science and Technology, ²Chiyoda Technol Corporation, ³Japan Atomic Energy Agency,)

Copper-67 is an attractive radionuclide for cancer therapy, because of emissions of medium energy beta particle (a mean energy of 141 keV) and gamma rays (185 keV) suitable for imaging, and its appropriate half-life (62 hours). However, the use of ^{67}Cu in clinical research has been limited due to the difficulty in obtaining a sufficient amount of ^{67}Cu . We have investigated the production of ^{67}Cu by the $^{68}\text{Zn}(n,x)^{67}\text{Cu}$ reaction ($x=n, p, d$) using fast neutrons from the $^{nat}\text{Be}(d,n)$ reaction at the Takasaki Ion Accelerators for Advanced Radiation Application. The separation and purification method of highly-purified ^{67}Cu which is capable for mouse experiments was established. In this study, the specific radioactivity of ^{67}Cu was determined by the dependence of DOTA concentration upon the labeling yield of ^{67}Cu -DOTA. The minimum DOTA concentration where the labeling efficiency is 100% is considered to be equal to the Cu concentration. The specific radioactivity of ^{67}Cu which is produced by irradiation of 5 g ^{68}ZnO for 24 hours with fast neutrons from 2 mA, 40 MeV deuterons + carbon, was estimated to be 2.1 GBq/ μg Cu at EOI.

P28 Examination of the isolation method of the radioactive microparticles in the environmental sample

TAGOMORI, H., ISHI, Y., KANAIZUKA, S., DOHI, T., IJIMA, K.
(JAEA)

In order to evaluate the chemical form and distribution of radionuclide in soil and litter near the Fukushima Daiichi Nuclear Power Plant, method for isolating radioactive particles were carried out. Main procedures for isolating radioactive particles are as follows; After drying and size classification, samples are decomposed with hydrogen peroxide. And then, IP method is carried out to monitor the distribution of radionuclide. A portion showing high radiation is selected as microscopic analysis which composed of Field Emission-Electron Probe Micro-Analyzer (FE-EPMA) and Energy-Dispersive Spectroscopy (EDS).

As results of above method, several particles with unique elemental compositions such as Ba was identified. Moreover, methods for isolating particles of 100 - 200 μm using the Micro Manipulator were established. The isolation of fine particles (less than 50 μm), were also applied by the meaning of digging a trench on the carbon tape using Focused Ion Beam (FIB).

P29 Search for the vacuum ultraviolet photons from $^{229\text{m}}\text{Th}$ reacted with HF gas

YASUDA, Y.¹, KASAMATSU, Y.¹, SHIGEKAWA, Y.¹, TAKAMIYA, K.², OHTSUKI, T.², MITSUGASHIRA, T.³,

SHINOHARA, A.¹

(¹Grad. Sch. Sci., Osaka Univ., ²Res. Reactor Inst., Kyoto Univ., ³The Oarai-branch, Inst. Materials Res., Tohoku Univ.)

Thorium-229m has the extremely low excitation energy, 7.8 ± 0.5 eV, which corresponds to the vacuum ultraviolet photon. It is expected that the decay mode of $^{229\text{m}}\text{Th}$ might change depending on its chemical condition: the internal conversion (IC) or gamma-ray transition. Recently, the IC electrons emitted from $^{229\text{m}}\text{Th}$ neutral atoms were reported. If we could detect the photons from $^{229\text{m}}\text{Th}$, direct observation of the change of the decay mode would be achieved. In the present study, we performed the photon detection for the sample prepared by reacting the highly-charged $^{229\text{m}}\text{Th}$ recoil ions with HF gas.

The ^{233}U electrodeposited sample was set in the Collection Apparatus for Recoil Products (CARP). Thorium-229m ions were collected in He or He/HF mixture gas atmosphere. To detect the photons, the photomultiplier tube which detects 4-10 eV photons was used. In the results of photon detection for the $^{229\text{m}}\text{Th}$ samples collected in the both He gas and He/HF mixture gas, the decaying photons were observed. Because the main component of these photons might be induced by β decays of daughter nuclides, the newly purified ^{233}U sample was prepared and set in CARP. In the case of using the new ^{233}U sample, no significant photons were detected. Thorium-229m ions were considered not to react with HF gas sufficiently.

P30 Determination of ^{125}Sb and ^{129}I in radioactive particles derived from FDNPP

MATSUO, K.¹, SUEKI, K.¹, SATOU, Y.², YOSHIKAWA, H.², MINOWA, H.³, SASA, K.¹, MATSUNAKA, T.⁴, MATSUMURA, M.¹, TAKAHASHI, T.¹, SAKAGUCHI, A.¹

(¹CRIED, Univ. of Tsukuba, ²JAEA, ³The Jikei University School of Medicine, ⁴Kanazawa Univ.)

In March 2011, radioactive particles were emitted into the environment from FDNPP. We wanted to presume how radionuclides were absorbed in particles when they generated and understand the process of generating radioactive particles. In this work, we tried to determine the amount of measurable radionuclides in these particles. We measured the radioactivity of gamma-ray of radioactive particles by Ge semiconductor detectors and determined the quantity of ^{125}Sb , ^{134}Cs and ^{137}Cs . And, we extracted iodine from the particles using the two experimental procedures and quantified the amount of ^{129}I by AMS. And then, the activity ratio of the target nuclide to ^{137}Cs on 2011/3/11 was calculated. The activity ratio of ^{134}Cs to ^{137}Cs was $0.912 \sim 0.935$ (Average : 0.918 ± 0.008). So, these particles were from the unit 1. And the average of activity ratio of ^{125}Sb to ^{137}Cs was $(1.1 \pm 0.4) \times 10^{-2}$ and it was comparable to the estimated radioactivity ratio. However, the activity ratio of ^{129}I to ^{137}Cs was $(1.1 \sim 5.8) \times 10^{-9}$ (Average : 2.9×10^{-9}) and it was smaller than the estimated radioactivity ratio by two orders of magnitude^[2]. From the results, it was presumed that the absorbed radionuclides of particles were different process at each radionuclide.

P31 Solvent extraction of Zr and Hf, lighter homologs of rutherfordium, with 2-furoyltrifluoroacetone

OOE, K.¹, KUSAKARI, S.², GOTO, S.¹, KUDO, H.², HABA, H.³, KOMORI, Y.³
(¹Grad. Sch. of Sci. and Tech., Niigata Univ., ²Fac. of Sci., Niigata Univ., ³RIKEN Nishina Center)

We have been investigating solvent extraction behavior of Zr and Hf using organic ligands such as 2-thenoyltrifluoroacetone (HTTA) for aqueous chemical studies of element 104, Rf. In the previous study, we found that it takes about several tens of minutes to attain extraction equilibrium of Zr and Hf with HTTA and another extractant which shows faster kinetics is required for chemical experiments with the short-lived ²⁶¹Rf. In this study, 2-furoyltrifluoroacetone (HFTA) was employed for solvent extraction of trace amounts of Zr and Hf. The extraction kinetics was investigated by a batch method and a rapid extraction apparatus using a flow injection analysis (FIA) technique. Radiotracers of ⁸⁸Zr and ¹⁷⁵Hf were produced using the AVF cyclotron at RIKEN. These radiotracers were chemically separated from target materials. Nitric acid solution containing the radiotracers and HFTA in toluene solution were used as aqueous and organic phases, respectively. In the batch extraction experiment, the extraction equilibrium of Zr and Hf with HFTA was attained in approximately 4 min, showing faster extraction kinetics than HTTA. In the presentation, the results of extraction experiments with the FIA apparatus will be also reported.

P32 The study on the method of the rapid screening analysis for alpha and beta nuclides in foods

YAMANAKA, J.¹, SHIMA, N.¹, TENNICHI, Y.¹, TAMARI, T.¹, MOMOSHIMA, N.¹
(¹Kyushu Environmental Evaluation Association)

The criterion value of radioactivity in foods is specified for radioactive cesium and the activity is conventionally measured by gamma spectrometry. However, analysis of alpha and beta nuclides in foods is not easy. We examined the screening method for Sr-90(Y-90), Pu and Am in foods. The nuclides were purified with a DGA resin and measured simultaneously using the α/β discrimination function of the low background liquid scintillation counter (LSC). The recoveries of Y-90, Pu-239+240 and Am-241 were more than 90% and almost all of disturbing elements, U, Th and Bi were effectively removed. Moreover, the optimum discrimination level of the α/β discrimination function was determined to be the minimum spillover value of α and β cross contamination. The detection lower limits were about 20 mBq at Y-90, and about 2 mBq at Pu-239+240 and at Am-241 for 100 minutes counting time using the optimum discrimination level. However, the incomplete removal of Po-210 is found to affect on the measurement in case of high Po contain samples.

P33 Reexamination of no-carrier-added purification of ²⁸Mg produced in the ²⁷Al(α ,3p) reaction.

KIKUNAGA, H.¹, KOMORI, Y.², HABA, H.², SHIBATA, S.², YANO, S.²
(¹ELPH, Tohoku Univ., ²RIKEN Nishina Center)

The isotope ²⁸Mg is useful in biological sciences as a radioactive tracer. Generally, ²⁸Mg is produced for each use in

nuclear reactions because of its short half-life of 21.6 h. The reaction ²⁷Al(α ,3p)²⁸Mg is often chosen for the production of ²⁸Mg owing to its potential to produce no-carrier-added ²⁸Mg tracer with a convenient and low-cost target. Several groups have reported separation methods of ²⁸Mg from Al targets based on adsorption, co-precipitation, solvent extraction, and the cation exchange column method. Although the cation exchange column method should yield a high purity tracer, it requires a large column and an eluant volume of a few hundred milliliters. In this work, we applied a co-precipitation method before the cation exchange column method to downsize the column volume. Downsizing chemistry leads to the reduction of waste and labor in separation procedures and the quality improvement of the tracer.

P34 Molecular structure effect on muon capture processes in benzene and cyclohexane

INAGAKI, M.¹, NINOMIYA, K.¹, NANBU, A.¹, KUDO, T.², TERADA, K.¹, SATO, A.¹, KAWASHIMA, Y.³, TOMONO, D.³, SHINOHARA, A.¹
(¹Grad. Sch. Sci., Osaka Univ., ²Sch. Sci., Osaka Univ., ³RCNP, Osaka Univ.)

Formation processes of muonic atoms are affected by the chemical environment of the muon-capturing atoms, as it is called chemical effect. Previously, we reported that the chemical effect on muon transfer process in benzene and cyclohexane in the gas phase was smaller than the detection limit. In this study, we performed muon irradiation experiment for liquid mixtures of benzene or cyclohexane and carbon tetrachloride to examine the chemical effect on muon transfer process in the liquid phase. The muon irradiation experiments were performed at the MuSIC-M1 beam line of the Research Center for Nuclear Physics (RCNP) in Osaka University. Muonic X-rays emitted after muon-capture for carbon or chlorine atoms were measured by high purity germanium detectors. Muon capture probabilities of carbon and chlorine atoms were determined from muonic X-ray spectra. We found that the muon capture probabilities of carbon atoms in cyclohexane samples were higher than that in benzene samples. This indicates that the muon capture probabilities of cyclohexane molecules are higher than those of benzene molecules.

P35 Improvement of detection sensitivity for long-lived radionuclide ³⁶Cl for the 6 MV tandem accelerator mass spectrometry system.

TAKANO, K.¹, SASA, K.¹, OHTA, Y.¹, TAKAHASHI, T.¹, HOSOYA, S.¹, OCHIAI, Y.¹, SUEKI, K.¹
(Univ. of Tsukuba¹)

The 6 MV tandem accelerator has been working on multi-nuclides Accelerator Mass Spectrometry (AMS) (¹⁰Be, ¹⁴C, ²⁶Al, ³⁶Cl, ⁴¹Ca, and ¹²⁹I) at the University of Tsukuba. AMS is highly sensitive measurement method for long-lived radionuclides. We can determine very low isotopic ratio (10^{-10} – 10^{-15}) in the sample by AMS, nevertheless if there is a small amount of measuring object. On the other hand, we have to care about the interference of isobar from the sample in AMS. For example, ³⁶S may cause serious interference on ³⁶Cl ($t_{1/2} = 3.0 \times 10^5$ yr)-AMS. ³⁶Cl is commonly used to determine the exposure age of

meteorites or rocks. The record of ^{36}Cl in the ice core has important information on the history of cosmic ray intensity over the past several hundred thousand years. However ^{36}Cl concentration in the Antarctic ice core is normally several thousand atoms g^{-1} . Analyses of $^{36}\text{Cl}/\text{Cl}$ values by accelerator mass spectrometry are estimated to 10^{-14} . Therefore, we should improve the method to remove ^{36}S for high sensitivity ^{36}Cl measurement for the 6 MV tandem accelerator mass spectrometry system. In this study, we tried to use AgBr as the backing material on sample holders. In addition, we made several AgBr materials under the different chemical processing conditions in order to compare the effect of ^{36}S elimination. We got the result that there is lower counts of ^{36}S under acid treating AgBr material as compared with the commercial AgBr reagent. In this presentation, we will introduce the result on the improvement of ^{36}Cl detection sensitivity for the 6 MV tandem accelerator mass spectrometry system.

P36 Development of a low energy atomic beam extraction technique for the determination of the valence electronic structure of Lawrencium ($Z = 103$)

TOMITSUKA, T.^{1, 2}, TOKOI, K.^{1, 3}, SATO, T. K.¹, ASAI, M.¹, TSUKADA, K.¹, TOYOSHIMA, A.¹, CHIERA, N. M.¹, KAMADA, H.^{1, 4}, NAGAME, Y.^{1, 3}, GOTO, S.².

(¹JAEA, ²Grad. Sch. of Sci. and Tech, Niigata Univ., ³Ibaraki Univ., ⁴Grad. Sch. of Engin., Nagoya Univ.)

In order to determine the valence electronic configuration of the ground state of Lawrencium (Lr, $Z = 103$), which is influenced by strong relativistic effects, we have developed a technique for production of a low energy atomic beam using a high-temperature nozzle method. To generate an atomic beam with a high efficiency, the generation rate and the directivity of atomic beam needs to be improved. Here the current status of the atomic beam source will be presented.

P37 Acceleration of analytical procedure for the Pu and Am analysis in soil using by ammonium salt fusion and solid phase extractions

YAMAMORI, K.¹, UESUGI, M.², YOKOYAMA, A.³

(¹ Graduate School of Natural Science and Technology, Kanazawa University, ²Advanced Science Research Center, Kanazawa University, ³Institute of Science and Engineering, Kanazawa University)

A rapid analysis is required to ensure safety in a nuclear accident. Accelerated procedures of the analysis for plutonium and americium were examined due to the demand. The dissolution of Pu and Am in soil was carried out by the ammonium salt fusing method, and the purification process was performed by using extraction disk or cartridge. In the dissolution-process, Pu and Am can be dissolved more quickly by the ammonium salt fusion than by the nitric acid leaching. Heating at 200-250 °C for 30 minutes is sufficient on a hot plate for the dissolution. The removal of organic substance was performed by addition of potassium permanganate solution. In the purification using a TEVA disk for Pu and using DGA cartridge and TEVA cartridge for Am. As a result, it was made possible to shorten the soil dissolution from 48 hours to 3 hours and the purification separation for Pu or Am from 12 hours

to 1 hour.

P38 Hydroxide precipitation experiment of alkaline earth metals for experiment of Nobelium

NAGASE, M.¹, KASAMATSU, Y.², KONDO, N.², SHINOHARA, A.²

(¹ Sci., Osaka Univ, ²Grad. Sch. Sci., Osaka Univ.)

Nobelium (No) belongs to actinide elements, and in the previous study, the most stable ion valency of No in aqueous solution is reported to be +2 although those of the other heavy actinide elements are all +3. However, the chemical experiments of No were very few because of the low production rates and short half-lives of the No isotopes. In the previous study, it was reported that the chemical behaviors of alkaline earth metals are similar to that of No. For the precipitation experiment of No, in this study, we investigated the hydroxide precipitation behaviors of alkaline earth metals (Mg, Ca and Ba) by applying the coprecipitation method with Sm hydroxide. We mixed 0.10 M of alkaline earth metal chloride in 0.20 M nitric acid and basic solutions (dil. and conc. NH_3 , 0.10 M, 1.0 M, 6.0 M or 12 M NaOH), and the prepared precipitate was filtered and separated from supernatant solution. The metal concentration of each solution was determined by titration with 0.010 M EDTA and the precipitation yield was obtained. The precipitation yields of the elements increased with the hydroxide ion concentration and the yields of heavier elements increased at higher hydroxide ion concentrations. These tendencies are consistent with the order of the solubility products of these elements.

P39 Alpha-electron coincidence measurement of ^{233}U for detecting the internal-conversion electron of $^{229\text{m}}\text{Th}$

SHIGEKAWA, Y.¹, KASAMATSU, Y.¹, YASUDA, Y.¹, SHINOHARA, A.¹, STELLMER, S.², KAZAKOV, G. A.², ROSECKER, V.², SCHUMM, T.²

(¹Grad. Sch. Sci., Osaka Univ., ²Institute for Atomic and Subatomic Physics, Vienna University of Technology)

The nuclide $^{229\text{m}}\text{Th}$ has extremely low excitation energy (7.8 ± 0.5 eV), which is close to the binding energy of the valence electrons. Therefore, it is estimated that the chemical environments affects the process of the nuclear decay of $^{229\text{m}}\text{Th}$ (internal conversion (IC) or gamma-ray emission) and also its half-life. Last year, it was reported that the IC electron originating from $^{229\text{m}}\text{Th}$ was detected for the first time and the half-life was determined to be around 7 μs . However, the excitation energy of $^{229\text{m}}\text{Th}$ has not been directly determined. In this study, we tried to detect IC electrons from $^{229\text{m}}\text{Th}$ recoiling out of ^{233}U with the alpha-electron coincidence measurement, and to determine the energy of the IC electrons with the retarding-field spectrometer for determining the excitation energy of $^{229\text{m}}\text{Th}$ directly. We prepared two types of ^{233}U sources on the Si detector, and measured alpha particles with the Si detector and electrons with the MCP detector placed above the ^{233}U source. IC electrons from $^{229\text{m}}\text{Th}$ should be detected about 7 μs after detection of alpha particles originating from ^{233}U decay. We were able to obtain some time spectra for the ^{233}U sources. As some delayed electrons were observed after alpha decay of ^{233}U and ^{232}U , IC electrons from $^{229\text{m}}\text{Th}$ have not yet been distinguished.

P40 Dissolved State of Radon with Cluster Molecules of Solvent

YAMADA, N.¹, SHIN, Y.¹, KAWASAKI, K.², YOKOYAMA, A.³

(¹ Grad. Sch. Nat. Sci. Tech., Kanazawa Univ., ^{2,3} Coll. and Inst. Sci. Eng., Kanazawa Univ.,)

Radon is a noble gas and the all isotopes of radon are radioactive. A method commonly used to measure radon in solution is liquid scintillation counting. This method measures radon extracted from a solution sample into scintillation cocktail (organic solvent). Radon dissolves well in water compared to other noble gases. The dissolved state of radon in water, however, remains undefined. The interaction between water molecules and a radon atom is important to solve the question. In 2001, existence of (H₂O)_n clusters (n=8-20) in solution were proposed by a theoretical calculation. In this study, enthalpies of noble gases (Ne, Ar, Kr, Xe, Rn) confined in (H₂O)₂₀ cluster were calculated with a density-functional-theory method and compared with standard enthalpies of solution of noble gases. As a result, enthalpy values of noble gases confined in the cluster (theoretical value) and those of solution (measured value) decrease with an increase of the atomic numbers of noble gases. In addition, it seems that enthalpies of solution correlate roughly with those of noble gases confined in the cluster. These results suggest dissolving of noble gas in water is related to the formation of cluster of water molecules with a noble gas atom.

P41 Observation of local fields in cobalt ferrite by means of nuclear spectroscopies

TAKENAKA, S.¹, SUGIMOTO, T.¹, SATO, W.^{1,2}

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

In our previous work, we measured hyperfine fields in magnetite (Fe₃O₄) and observed that ¹¹¹In (→¹¹¹Cd) atoms partly migrate from the tetrahedral *A* sites to the octahedral *B* sites at high temperature. For further understanding of this dynamic phenomenon, we extended our study to another spinel ferrite, Co_xFe_{3-x}O₄, and investigated their local fields by means of perturbed angular correlation (PAC) spectroscopy with the ¹¹¹In(→¹¹¹Cd) probe and Mössbauer spectroscopy with ⁵⁷Co(→⁵⁷Fe) probe. As a result, two different components having hyperfine magnetic interactions were observed in PAC spectra below *T_C*: one exhibits small distribution in the Larmor frequency assignable to the probe nuclei substituting for a cation site of inner part of the grains (“core”), and the other fraction to the probe nuclei at the surface and/or boundary of the grains (“shell”). As the cobalt concentration *x* increased, the magnetic hyperfine field at ¹¹¹In(→¹¹¹Cd) probe in the core region decreased and its distribution width increased. This result was compared with that obtained by the Mössbauer measurements. In the session, the Co concentration and temperature dependence of the PAC and Mössbauer spectra are discussed.

P42 Chemical environmental effect on muon capture between

[Fe(II)(CN)₆] and [Fe(III)(CN)₆]

NINOMIYA, K.¹, KUBO, K.², KITANAKA, M.¹, TAMPO, M.³, KAWAMURA, N.³, STRASSER, P.³, MIYAKE, Y.³, SHINOHARA, A.¹

(¹Grad. Sch. Sci., Osaka Univ., ²ICU, ³KEK)

A muonic atom is an atomic system that has one negatively charged muon instead of an atomic electron. In muonic atom formation, chemical environmental effect has been known. Muon capture probability and initial muon quantum level are strongly influenced by the chemical environment of muon capturing atom. In this work, we focused on the muon capture process for iron complexes to investigate chemical environmental effect. Muon irradiation experiment were performed at D2-experimental area in J-PARC/MUSE. Muonic X-rays emitted after muonic atom formation were measured by HPGe detectors for [Fe(II)(CN)₆] and [Fe(III)(CN)₆] samples with incident muon momentum of 30 MeV/c. From muonic X-ray intensities, we will discuss initial state of captured muon for K, Fe, C and N atoms for each sample.

P43 Electronic state conversions of iron complexes with Brønsted acid and base

SAIKI, R., AKIYAMA, L., SHIGA, T., OSHIO, H.

(Univ. of Tsukuba)

The electronic state of bistable complexes can be changed by various external stimuli such as heat, light, and pressure. However, research on control of spin state by deprotonation is still limited. In this work, the electronic states of a series of new mononuclear iron complexes with a Brønsted acid ligand H₂L were investigated. Based on deprotonation and oxidation conditions, four kinds of bis-chelate complexes were synthesized and their magnetic and electrochemical properties were studied. The Electronic state of the iron ions in all complexes was determined by single crystal XRD, magnetic, and Mössbauer measurements. [Fe^{II}(H₂L)₂](BF₄)₂ and doubly deprotonated neutral complex [Fe^{II}(HL)₂] show spin crossover behavior at 260 K and 350 K, respectively. This difference in transition temperature originates from the change of ligand field strength by deprotonation. On the other hand, triply deprotonated neutral complex [Fe^{III}(L)(HL)] and fully deprotonated {(C₂H₅)₄N}[Fe^{III}(L)₂] contain Fe(III) in the low spin state, suggesting further deprotonation of ligand stabilizes the high oxidation state.

P44 Ion exchange behavior of Zr, Hf, and Th with ion-exchange fiber –Basic study for ion-exchange experiments of Rf using ion-exchange fiber–

YOKOKITA, T.¹, YANO, S.¹, KOMORI, Y.¹, HABA, H.¹

(¹RIKEN)

To investigate applicability of ion-exchange fiber for ion-exchange experiments of Rf, we performed ion-exchange experiments of Zr, Hf (homologues of Rf), and Th (pseudo homologue of Rf) with anion- and cation-exchange fibers. Distribution ratios (*Q_d*) of ⁸⁸Zr, ¹⁷⁵Hf, and ²³⁴Th on the anion-exchange fiber (Nichibi, IEF-BrA-SA) and cation-exchange fiber (Nichibi, IEF-MR-SC) were measured in 0.1–1 M H₂SO₄ by batch method, and were compared with those of the

anion-exchange resin (Mitsubishi Chemical, MCI GEL CA08Y) and the cation exchange resin (Mitsubishi Chemical, MCI GEL CK08Y), respectively. In the anion exchange, it was found that the Q_a values of Zr and Hf attain to constants at the shaking times of 1800 s for the anion-exchange fiber and of 10 s for the resin, indicating that the ion-exchange reaction with the resin is shorter than that with the fiber. In the cation exchange, we also found that the time required for the equilibrium with the resin is faster than that with the fiber. From these results, we concluded that the ion-exchange fibers used in this work are unavailable for the experiments with the short-lived ^{261}Rf ($T_{1/2} = 68$ s). However, we observed the fast equilibrium with the anion-exchange resin in 0.1–1 M H_2SO_4 . This system would be applicable for the ion-exchange experiments with ^{261}Rf .

P45 Application of triple quadrupole ICP-MS(ICP-MS/MS) to radiochemistry

SHIKAMORI, Y.¹, NAKANO, K.¹, WOODS, G.¹, LIBA, A.¹, SAKAGUCHI, A.²

(¹Agilent Technologies ²CRiED, Univ. of Tsukuba)

Trace measurement of ^{237}Np by conventional inductively coupled plasma mass spectrometry is hindered by the presence of uranium within the sample, the potential interference comes from peak broadening (abundance sensitivity interference) of the adjacent ^{238}U isotope. Furthermore, minor but still important polyatomic interferences can be present from the hydrides of lighter U isotopes – $^{235}\text{UH}_2$; $^{235}\text{U}^2\text{D}$; $^{234}\text{U}^1\text{H}^2\text{D}$.

Agilent 8900 ICP-QQQ(ICP-MS/MS) was used in this work, which features axial acceleration across the collision reaction cell which gives a higher product ion yield when using reaction chemistry within the cell.

To test the U-based interference removal, Np-237 was spiked into a 10ppm U matrix solution at ppt level. Np-237 was measured under MS/MS conditions both on-mass and mass-shift using O_2 reaction gas to produce the NpO_2 product ion.

When operating under MS/MS mode, the peak overlap is eliminated by improving abundance sensitivity from $\sim 10^{-7}$ to $\ll 10^{-10}$. LOD and BEC using MS/MS O_2 reaction mode and measuring as NpO_2 improved the LOD to 0.56ppq and BEC to 0.32ppq.

We will report analytical figures of merit achieved and details of the optimized method with some of back ground data for the optimization.

P46 Effect of dissolved ozone on adsorption-desorption mechanisms of various types of radionuclides using tannic acid-type organic composite adsorbent

TACHIBANA, Y.¹, HASHIMOTO, S.¹, OKUMURA, S.², NOGAMI, M.³, SUZUKI, T.¹, NOMURA, M.², KANESHIKI, T.²
(¹Dept. of Nuclear System Safety Engineering, Nagaoka Univ. of Technol., ²Lab. for Advanced Nuclear Energy, Tokyo Tech, ³Dept. of Electric and Electronic Engineering, Kindai Univ.)

We have focused the excellent adsorption ability of tannic acid for many kinds of metal ions in aqueous solutions and have successfully synthesized novel ion-exchange resins chemically combined with hydrolyzed tannic acid and typical anion-exchange

resins, named improved tannic acid-type adsorbents (ITAs). In our present research, the adsorption behavior on the synthesized ITAs of manganese, cobalt, strontium, yttrium, iodine including iodide species, and the like, which has the possibility released into our living environment after nuclear severe accidents, have been studied in several types of river water at a temperature range of 278–333 K and in a wide pH range, compared with those of some resins before and after ozonation. We have already confirmed that ITAs can strongly adsorb these species, which should be separated completely and little negative adsorption behavior of main species such as sodium, potassium, calcium, magnesium is observed in river water. Moreover, the thermodynamic parameters have been obtained and the differences of adsorption mechanisms between the radionuclides and ITAs have been discussed. In order to evaluate the potential application of ITAs for our novel ozone water purification system, the chromatographic experiments of the radionuclides using ITAs have also been performed in river water at 298 K.

P47 Summary of reports on environmental radioactivity effected from the accident at the TEPCO Fukushima Daiichi nuclear power plant

BESSHO, K.¹, MATSUMURA, H.¹, MIURA, T.¹, IIMOTO, T.², KINOSHITA, N.³, SAKAGUCHI, A.⁴, SUGIHARA, S.⁵, TAKAMIYA, K.⁶, TAGAMI, K.⁷, NAGAO, S.⁸, YASUDA, K.⁹

(¹KEK, ²Univ. Tokyo, ³Shimizu Corp., ⁴Univ. Tsukuba, ⁵Kyushu Univ., ⁶Kyoto Univ., ⁷QST-NIRS, ⁸Kanazawa Univ., ⁹JAEA)

Workshop on Environmental Radioactivity was established in 2000 and has been held every year on February or March at KEK, Tsukuba. [Host: 1) Radiation Science Center, KEK; 2) Subcommittee on α -emitters and Environmental Radioactivity, JNRS; Co-host: 1) Subcommittee on Health Physics and Environment Science, Atomic Energy Society of Japan, 2) The Japanese Radiation Research Society, 3) Japanese Society of Radiation Safety Management.] The proceedings with peer review has been published after every workshop.

After the accident at the TEPCO Fukushima Daiichi nuclear power plant occurred in 2011, a large numbers of scientific reports related to the accident were presented in this workshop. Researchers, technicians, local government staff, and general citizens have participated in the workshop to exchange information on various subjects. The organizing members of the workshop summarized 326 reports in the workshop during 2012 to 2016 and related publications. The summary report was published as follows.

KEK Report 2016-3 (ISBN978-4-9907232-9-3)

“Summary of study on environmental radioactivity effected from the accident at the TEPCO Fukushima Daiichi nuclear power plant in 5 years after the accident” (in Japanese)

<https://lib-extopc.kek.jp/preprints/PDF/2016/1624/1624003.pdf>

本年会・討論会の開催にあたりましては、
以下の企業、団体からご協力をいただきました。
厚く感謝申し上げます。

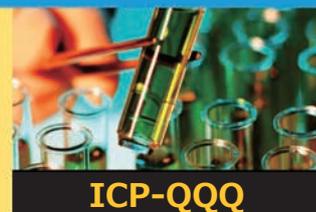
協賛

サンゴバン（株）
東京ニュークリアサービス（株） TNS
長瀬ランダウア（株）
富士電機（株）

広告

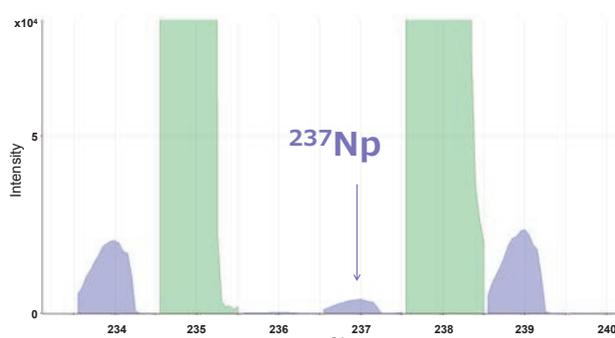
Agilent（株）
北原商事（株）
サンゴバン（株）
桑和貿易（株）
東京ニュークリアサービス（株） TNS
長瀬ランダウア（株）
富士電機（株）
ミリオンテクノロジーズ・キャンベラ（株）
（株）アトックス
（一財）九州環境管理協会
（株）千代田テクノル
日本アドバンストテクノロジー（株）
（株）日本環境調査研究所

Agilent 8900 トリプル四重極ICP-MS ICP-MS/MS



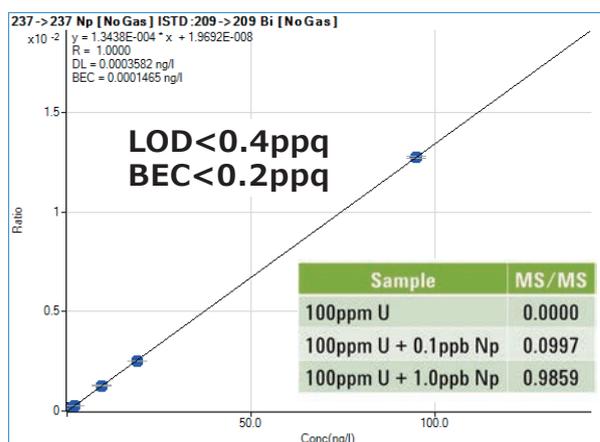
放射性元素分析へのICP-MS/MSの応用例

ICP-MS/MSによる²³⁷Npの分析



10mg/L中 0.1μg/L Npのスペクトル

※ウランのテーリングの影響を受けにくい



100ppm U にNpを標準添加した検量線



濃度と同位体情報を同時取得

Agilent 8900 トリプル四重極 ICP-MS

アジレント・テクノロジー株式会社

本社/〒192-8510 東京都八王子市高倉町9-1

●カスタムコンタクトセンタ 0120-477-111

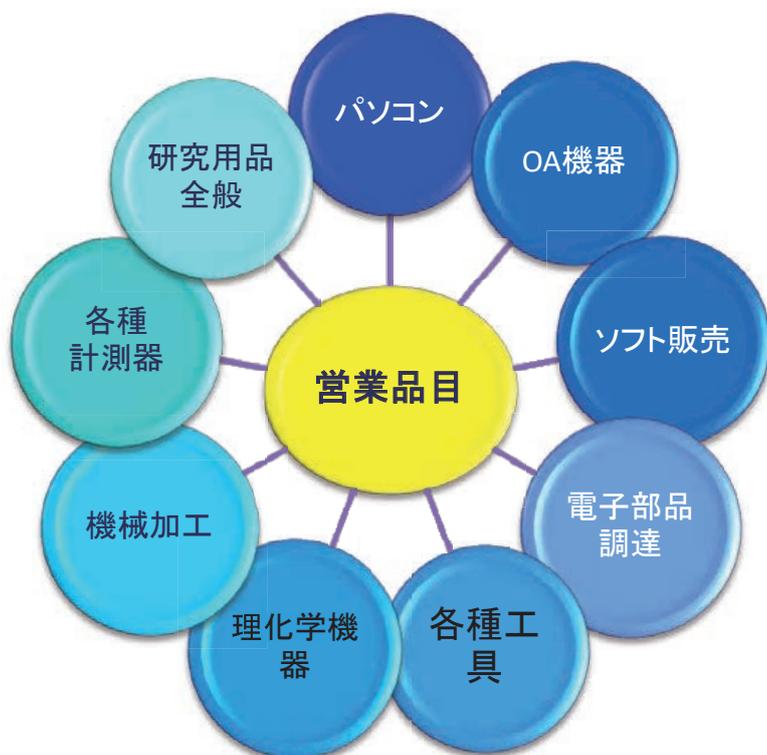
www.agilent.com/chem/jp

The Measure of Confidence



Agilent Technologies

研究のお役に立つ商品を扱う総合商社です。
まずはお気軽にお問い合わせ下さい。



主な取引先

新日鐵住金株式会社 技術開発本部
新日鐵住金株式会社 君津製鐵所
株式会社本田技術研究所
株式会社 ホンダ・レーシング
筑波大学
東京大学
高エネルギー加速器研究機構
産業技術総合研究所
物質材料研究機構
理化学研究所



北原商事株式会社

本社

〒177-0051

東京都練馬区西大泉6-1-22

03-6904-6961

03-6904-6981

つくば営業所

〒305-0023

茨城県つくば市上ノ室2228-7

029-863-0002

029-863-0003

富津営業所

〒293-0012

千葉県富津市青木1536-3

0439-87-5247

0439-87-5517

とちぎ営業所

〒321-3426

栃木県芳賀郡市貝町赤羽3525-48

0285-67-0078

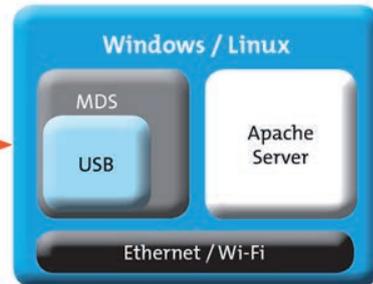
0285-67-0079



サンゴバンは、素材からフローブ・特殊用途まで、グローバル・スタンダードな世界品質のシンチレータを皆様に提供いたします。

Sensor-Kit MCA-BASE

Scintillation Detectors
(NaI,LaBr₃,BGO,Plastic,Liquid,・・・)
+Divider + MCA + HV are only in
One-body or Two-bodies



Windows Client

Android Client

Linux Client

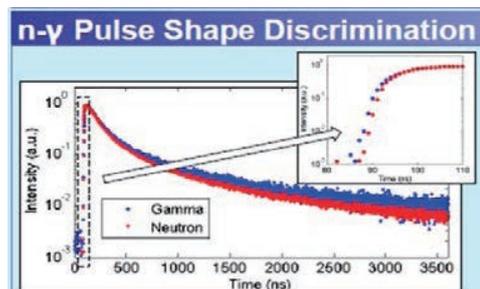
売れてます!

LaBr₃ (Ce)



FWHM=2.8% @662keV
Decay Time 16 nsec

中性子用シンチレータ Cs₂LiLaBr₆ (Ce³⁺) [CLLB:Ce³⁺]



サンゴバン株式会社

〒102-0083 東京都千代田区麴町 3-7
Tel 03-3263-0559 Fax 03-5212-2196

URL <http://www.crystals.saint-gobain.com/>

お問合せ yasuo.watanabe@saint-gobain.com

(担当 渡辺)

フィンランド国

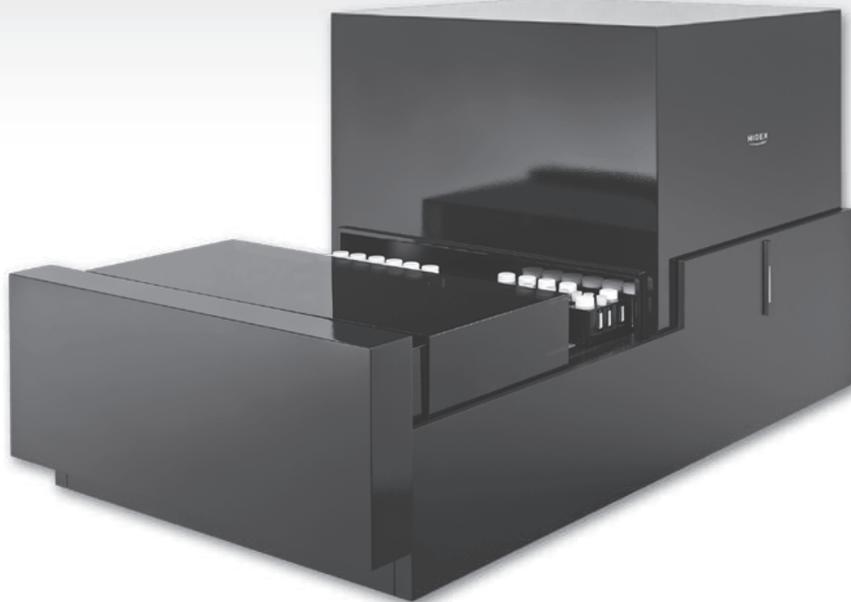
HIDEX

Hidex600SL型 世界唯一のTDCR技法搭載 全自動液体シンチレーションカウンター

世界初で、唯一の3本の光電子増倍管を駆使したTDCR技法により、今やRI線源を内蔵したクエンチング補正のための外部標準線源機構は、全く不要となりました。このため、測定時間の短縮化はもちろんのこと、ケミカル及びカラーのクエンチングも、たった1本の補正カーブで正確、また迅速に行え、正確なBq値が得られるようになりました。

更に、最新の“デジタル鉛シールド機構”の採用により、バックグラウンドを平均30%も低減でき、加えて“ルミネッセンス フリーモード機構”の使用により、ルミネッセンスを一切気にせず、測定が可能となります。また、⁹⁰Srの測定の場合、放射平衡（10～14日）を待たずにTDCR技法を用いてチェレンコフ測定（シンチレータを使用しない）により、簡単にクエンチングのモニターを行いながら、サンプル調整後、直ちに測定可能です。

今やドイツを中心に、欧米各国より“TDCR技法”がいかに優れているかに関する科学文献が60題以上も発表されています。当社のウェブサイト到现在16題掲載しております。



上記の他、従来の300SL型、そして極低レベルの環境測定（³H水で1Bq/L）ができるスーパー低レベル液シン（300SL/SLL型）と、多サンプルも装填使用出来る600SL/SLL型もございます。

主な納入先

放射線医学総合研究所
筑波大学大学院
国立海洋研究開発機構
東京慈恵会医科大学

国立保健医療科学院
産業技術総合研究所
九州大学RIセンター
米国国立衛生研究所(NIH)

東京工業大学 先導原子力研究所
大阪医科大学
国際原子力機構(IAEA)
フランス原子力エネルギー庁



桑和貿易株式会社

E-mail : office@sowa-trading.co.jp

URL : http://www.sowa-trading.co.jp

桑和貿易

検索

東京：〒101-0032 東京都千代田区岩本町1丁目7番1号（瀬木ビル）
TEL.03(3862)2700(代) / FAX.03(3862)6300

大阪：〒532-0011 大阪市淀川区西中島5丁目3番4号（新大阪高光ビル）
TEL.06(6390)2151(代) / FAX.06(6390)5620

TNS

お悩みではありませんか？

- 放射線安全管理を一元管理したい！
- 校正していないサーベイメーターの測定値が正しいか知りたい！
- 施設を変更許可申請したいが業務が煩雑、代行して欲しい！
- PET用サイクロなど加速器施設の計算および実測の放射化評価がしたい！
- 放射線施設を廃止したい！
- お米、果物、飲料水などの放射能濃度が知りたい！
- RIを使用した化学実験を代行して欲しい！
- 放射線に係わる業務について困った！

そのお悩みTNSが解決いたします

安全設計・評価

- ◆ 施設設計
- ◆ 遮蔽設計
- ◆ 安全評価
- ◆ RI施設の許認可申請業務代行

施設の管理・運営

- ◆ 大規模施設の運用管理
- ◆ 放射線管理

分析・測定・校正サービス

- ◆ 放射能分析
- ◆ 現地における放射線測定
- ◆ サーベイメータの確認校正

受託試験研究

- ◆ 環境物質の分析、挙動解析
- ◆ トレーサー試験
- ◆ 解体廃棄物の物理特性試験

研究及び技術開発サポート

- ◆ 研究サポート
- ◆ 技術開発サポート

保守点検・工事

- ◆ 施設の保守・点検
- ◆ 施設の改造、解体工事

各種機器販売

- ◆ 放射線管理区域の空調機器の販売
- ◆ 放射線管理区域用機器の製造・販売

Luminess

豊富なデザイン&高品質サービス

ルミネスバッジ サービス

個人被ばく線量測定



 長瀬ランドウア株式会社

本社/〒300-2686 茨城県つくば市諏訪C22街区1 Tel.029-839-3322 Fax.029-836-8441
大阪営業所/〒550-8668 大阪市西区新町1-1-17 Tel.06-6535-2675 Fax.06-6541-0931
ホームページアドレス/ <http://www.nagase-landauer.co.jp/>

富士電機の サーベイメータ・測定器

富士電機では、RI施設の管理を容易に実現する放射線管理システムをはじめ、放射線管理における様々な用途に応じた測定器類を取り揃えています。

X線・γ線の線量測定

電離箱サーベイメータ
NHA1



ハンディサーベイメータ
NHE



シンチレーションサーベイメータ
NHC 7



中性子の線量測定

中性子サーベイメータ
高感度タイプ
NSN2



中性子サーベイメータ
軽量タイプ
NSN3



β/γ線汚染測定



GMサーベイメータ
NHJ120

α/β線汚染測定



半導体サーベイメータ
NHJ2

環境線量の定点測定



環境線量計システム
NSDシリーズ

個人被ばく測定



電子式個人線量計
DOSEiシリーズ

富士電機株式会社
放射線システム部

〒191-8502 東京都日野市富士町1番地
ホームページURL <http://www.fujielectric.co.jp>
お問合せ先アドレス fric-info@fujielectric.com

北海道 011-221-5482
東北 022-716-0203

東京 042-585-6024
中部 052-746-1032

関西 06-7166-7346
九州 092-262-7844



MIRION
TECHNOLOGIES

ミリオンテクノロジーズ・キャンベラの放射線計測システム



電気冷却式 液体窒素循環型クライオスタット
Cryo-Cycle™ II
(クライオサイクル II)



電気冷却式 クライオスタット
Cryo-Pulse® 5 plus
(クライオパルス 5 プラス)



ゲルマニウム半導体検出器



DSA-LX™ :
デジタルシグナルアナライザ



ISOCSTM :
ガンマ線分析システム



InSpector 1000™ : 可搬型
デジタルスペクトロサーベイメータ



5XLBシリーズ:
低バックグラウンド α/β 自動計測システム



LB4200 型:
多チャンネル低バックグラウンド
 α/β 計測システム



ガンマアナリスト:
サンプルチェンジャー付
ガンマ線自動分析システム

放射線 安全性 さらなる水準へ

CANBERRA

ミリオンテクノロジーズ・キャンベラ株式会社
東京本社 TEL 03-5835-5402 大阪営業所 TEL 06-4806-5662
www.canberra.com/jp/ E-mail jp-sales@mirion.com

国内外の叡智をたずさえ、 原子力産業再生の最前線へ。

社会と産業を支えるクリーンエネルギー原子力。
アトックスは、その安全と安定した運転に欠かせないさまざまな
メンテナンス事業を展開しています。

原子力発電所、原子燃料サイクル施設、ラジオアイソトープ (RI) 事業所などを対象に
放射性汚染除去、産廃物処理、放射線管理施設の保守・補修業務をはじめ
質の高いトータルメンテナンスを提供しています。

アトックスはこれからも、人と地球を見つめ、
安全・清潔・便利さを追求し続けます。

人×技術でNext Stageへ



株式会社 アトックス

業務内容

■福島復興事業

- 放射線管理 ●環境修復 ●除染作業 ●滞留水処理
- 下水処理 ●廃棄物処理

■原子力発電関連施設・原子燃料サイクル施設のトータルメンテナンス

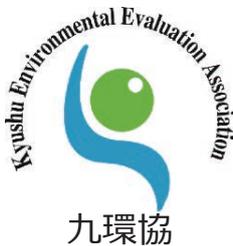
- 放射線管理 ●放射性汚染除染 ●廃棄物処理 ●ランドリ
- 清掃・区域管理 ●設備／装置運転・点検保守 ●輸送
- 分析 ●各種工事

■RI使用施設関連業務及び医療事業関連業務

- 放射線管理 ●放射性汚染除去 ●廃棄物処理補助
- 管理区域内清掃 ●施設／設備運転・点検保守 ●加速器運転
- フィルター交換 ●デコミッション・設備工事
- PET治験薬受託製造 ●放射性薬剤取扱業務
- 放射性注射剤製造施設の構築・点検保守 ●同位体販売

- 本 社：〒108-0014 東京都港区芝4-11-3 芝フロントビル
TEL.(03)6758-9000 FAX.(03)3453-3821
- 事業開発部：〒108-0014 東京都港区芝4-11-3 芝フロントビル
TEL.(03)6758-9004 FAX.(03)3453-3821
- 技術開発センター：〒277-0861 千葉県柏市高田1201
TEL.(04)7145-3330 FAX.(04)7145-3649
- RI・医療事業部署：東海営業所 TEL.(029)282-1662
大洗営業所 TEL.(029)266-1331
東京営業所 TEL.(04)7141-1321
大阪営業所 TEL.(06)6384-6730
神戸医療事業オフィス TEL.(078)599-6851
玄海事業所 TEL.(0955)52-3241

URL: <http://www.atox.co.jp/> (本社) <http://www.atox-isotope.jp/> (同位体販売) <http://www.medical-atox.com/> (医療事業)



九環協

調査、分析、評価まで
一連の業務を総合的にサポート

一般財団法人 九州環境管理協会



★業務内容★

- 環境放射能分析：放射性セシウム、放射性ストロンチウム
トリチウム、プルトニウム等
放射能分析全般
- 調査分析：河川・海洋調査、植生・動物調査、生態系調査
生物分類・同定、大気・水質・土壌分析
ダイオキシン類分析等
- 予測解析：環境アセスメント、数値シミュレーション等
- 計画設計：環境基本計画、地球温暖化防止計画等
- 啓発事業：環境教育等

【本部】 〒813-0004 福岡市東区松香台 1-10-1
TEL: (092)662-0964 (調査分析部), 問合せmail: syougai@keea.or.jp
ホームページ: <http://www.keea.or.jp>

【東京事務所】 〒105-0001 東京都港区虎ノ門 1-8-7 富田屋ビル7F
TEL: (03)6257-3028



千代田テクノルは
放射線

を 測る から 守る
で 治す

放射線は危険な性質を持っている反面、有効に利用すれば人類に大きなメリットを与えてくれる無限の可能性をそなえています。

千代田テクノルは、医療・原子力・産業・放射線測定などの各分野において、放射線を安全に有効利用するための機器やサービスをトータルに提供。

放射線の「利用」と「防護」の双方において、お客様のあらゆるニーズにきめ細かく対応しています。

株式会社 **千代田テクノル**

U R L : <http://www.c-technol.co.jp>

e-mail : ctc-master@c-technol.co.jp

千代田テクノル



JQA-QM8513
Tokyo・Osaka
Kashiwazaki Karifu



日本アドバンステクノロジー株式会社

「お客様のニーズを技術で支援する」 先端科学専門技術者集団の会社です

核融合分野

JT-60SA関係
ITER関係
IFMIF関係等

加速器分野

J-PARC加速器関係
MLF関係
STF関係等

レーザー分野

J-KAREN関係
激光XII関係
立命館SR関係等

原子力分野

HTTR関係等

設計・製作
工事

機器製作
据付工事

まずはご連絡ください

TEL

本社:029-283-3155 営業部:029-283-3193

住所

〒319-1112 茨城県那珂郡東海村村松字平原3129番地45

URL

<http://www.nat-web.com/>

日本アドバンステクノロジー



ラジオアイソトープ (RI) 取扱施設

Radioisotope(RI) handling facility

医療放射線 (核医学検査、放射線治療) 取扱施設

Medical radioactive rays handling facility

加速器取扱施設

Accelerator handling facility



技術開発研究所 (埼玉県吉川市)

● 放射線関係法令に基づく放射線モニタリング

- ・作業環境モニタリング
線量当量率測定
放射性表面汚染密度測定
空気中の放射性物質濃度測定
水中の放射性物質濃度測定
- ・個人モニタリング
放射線被ばく管理
- ・施設周辺の環境モニタリング
環境試料の放射能測定

● RI 取扱施設および機械装置等の保守管理業務

● RI 取扱施設設計・施工・コンサルティング

● RI 施設廃止工事

● 放射線関係法令に基づく各種申請書作成

● 放射線管理関連業務

● 施設管理コンサルタント業務

● 放射線業務従事者教育訓練

● 高度医療機器販売

● RI 管理関連ソフト開発、遮蔽用具・什器商品開発等

<http://www.jer.co.jp/>

株式会社 **日本環境調査研究所**
青森 仙台 柏崎 福島 茨城 東京 横浜 名古屋 大阪

作業環境測定機関登録 11-4 (放射性物質)

管工事業 / とび・土工事業 / 機械器具設置工事業 / 建具工事業

ISO9001 認証

技術開発研究所 〒342-0008 埼玉県吉川市旭 8-3
本 社 〒160-0023 東京都新宿区西新宿 6-24-1

Tel. 048-991-9461 Fax. 048-991-9460
Tel. 03-5322-2271 Fax. 03-5322-2272

**2017 日本放射化学会年会・第 61 回放射化学討論会
研究発表要旨集**

編集・発行：2017 日本放射化学会年会・第 61 回放射化学討論会
実行委員会（委員長：末木 啓介）

事務局： 筑波大学アイソトープ環境動態研究センター
〒305-8577 つくば市天王台 1-1-1
E-mail: sorc61@ied.tsukuba.ac.jp

発行日： 2017 年 9 月 1 日
印刷： 松枝印刷株式会社

※ 許諾・許可なしに本要旨集の全部もしくは一部の
転載、翻訳、複写、その他の複製を禁じます。

© 2017 日本放射化学会