1A01 Spontaneous fission measurements for nuclei around neutron-rich Fm region produced by using an ^{254}Es target

ASAI, M.[1], TSUKADA, K.[1], HIROSE, K.[1], TOYOSHIMA, A.[1], TOMITSUKA, T.[1], CHIERA, N. M.[1], ITO, Y.[1], MAKII, H.[1], NAGAME, Y.[1], NISHIO, K.[1], ORLANDI, R.[1], RATHA, K. K.[1], SATO, T. K.[1], SUZUKI, H.[1], TOKOI, K.[1], VERMEULEN, M. J.[1], YANAGIHARA, R.[1], NISHINAKA, I.[2], KAMADA, H.[3], SHIBATA, M.[3], GOTO, S.[4], HIROSE, H.[4], HAYASHI, H.[5], SAKAMA, M.[6]
([1]JAEA, [2]QST, [3]Nagoya Univ., [4]Niigata Univ., [5]Kanazawa Univ., [6]Tokushima Univ.)

Spontaneous fissions of the nuclei around neutron-rich Fm region, ^{256}Fm, ^{258}Fm, and ^{259}Lr, have been measured using an ^{254}Es target at the JAEA Tandem Accelerator Facility. These three nuclei show different fission properties: the asymmetric fission, the high total-kinetic-energy (TKE) symmetric fission, and the low TKE symmetric fission. The mechanisms of these three fissions should be very different, and it is of great interest to elucidate how these differences occur. We have produced ^{256}Fm and ^{258}Fm in multinucleon-transfer reactions with a ^{254}Es target and ^{18}O projectiles, and produced ^{259}Lr in the ^{248}Cm(^{15}N,4n) fusion-evaporation reaction. The reaction products were mass-separated with a gas-jet-coupled on-line isotope separator, and were delivered into a rotating wheel alpha- and fission-fragment detection system equipped with 4 pairs of Si detectors, where the fission fragments were measured in coincidence. By comparing the mass and TKE distributions deduced from the present experimental data, we will discuss the differences in fission mechanism among ^{256}Fm, ^{258}Fm, and ^{259}Lr.

1A02 Formation and thermochemical properties of oxychlorides of Nb and Ta - Towards the gas-phase investigation of the Superheavy Element dubnium (Db) oxychloride.

CHIERA, N. M.[1]、SATO, T. K.[1]、ASAI, M.[1]、SUZUKI, H.[1,2]、TOKOI, K.[1,2]、TOMITSUKA, T.[1,3]、TOYOSHIMA, A.[1]、TSUKADA, K.[1], NAGAME, Y.[1,2]
([1]JAEA, [2]Ibaraki Univ., [3]Niigata Univ.)

The adsorption of single molecules (i.e., tracer-amount scale) of $NbOCl_3$ and $TaOCl_3$ on quartz surfaces was explored by applying an isothermal gas-chromatographic method. Thermochemical information was obtained, in good agreement with literature data. The future accomplishment of comparative studies with $DbOCl_3$ under the same experimental conditions will provide a direct information on the volatility trend in Group-5 elements, and hence, on the magnitude of relativistic effects on dubnium’s electronic structure.

1A03 Development of a slow atomic beam source for Superheavy elements

TOMITSUKA, T.[1,2], TOKOI, K.[1,3], SATO, T. K.[1], ITO, Y.[1], SUZUKI, H.[1,4], ASAI, M.[1], TSUKADA, K.[1], TOYOSHIMA, A.[1], CHIERA, N. M.[1], GOTO, S.[2], NAGAME, Y.[1,3]

([1]ASRC, JAEA, [2]Graduate School of Science and Technology, Niigata Univ., [3]Graduate School of Science and Engineering, Ibaraki Univ., [4]College of Science, Ibaraki Univ.)

In order to experimentally determine the ground state electronic configurations of Superheavy element atoms, we have developed a slow Atomic Beam Source (ABS) using a high-temperature nozzle type cavity. In this study, we conducted an off-line experiment using stable isotopes of Cd to examine the extraction of the Cd beams. A symmetrical beam distribution with respect to the atomic beam edge was obtained. The obtained results, together with Monte-Carlo simulations for atomic beam profiles, will be presented.

1A04 The Study on Carbonyl Complexes of Group VII Elements

WANG, Y.[1,2], QIN, Z.[2], FAN, F.[2], CAO, S.[2], ZHANG, J.[2], YANG, J.[2], HABA, H.[1], KOMORI, Y.[1], YOKOKITA, T.[1], MORIMOTO, K.[1], KAJI, D.[1]

([1] Nishina Center for Accelerator-Based Science, RIKEN, [2]Institute of Modern Physics, Chinese Academy of Sciences)

Towards chemical characterization of the superheavy element bohrium (Bh), experiments with lighter its homologues Tc and Re are necessary to be investigated in advance. Online gas-phase chemistry study with single-atom quantities of Tc and Re carbonyls were carried out at RIKEN and IMP. Short-lived Tc and Re isotopes were produced via the spontaneous fission of ^{252}Cf and the ^{nat}Gd(^{23}Na,xn)$^{172-177}$Re reactions, respectively. Volatile carbonyls of Tc and Re were synthesized and their gas-phase properties were systematically studied using an online low temperature isothermal chromatography. For both Tc and Re carbonyls, the adsorption enthalpies on the Teflon surface were about $\Delta H_{ads} = -43\pm1$ kJ/mol, and the highest relative chemical yields of about 25% were measured. Moreover, the laser-ablation time-of-flight mass-spectrometry technique was employed to deduce the species of the Re carbonyls formed with a small amount of stable Re atoms and CO gas in the gas phase. Combined with density functional theory calculations, for mononuclear Re carbonyls, the most stable species were suggested to be the carbonyls with the coordination number of 6 and 5 for Re^+ and Re neutral atoms, respectively.

1A06 Hydration Structure of Einsteinium by Synchrotron Radiation Based Analysis

YAITA, T.[1], SUZUKI, S.[1], KOBAYASHI, T.[1], SHIWAKU, H.[1], DOI, R.[1], YAMAGAMI, H.[2], TSUKADAS, K.[3], TOYOSHIMA, A.[3], ASAI, M.[3]
([1]JAEA-MSRC, [2]Kyotosngyo Univ., [3]JAEA-ASRC)

The ionic radii of trivalent actinide and lanthanide systematically shrink with an increase in atomic number. This tendency, namely Actinide or Lanthanide Contraction", have been observed by the interatomic distances in oxide, the hydration bond distance and so on. Especially, a breaking point due to hydration number change often appears in lanthanide solution system. In contrast, regarding actinides, such a breaking point has not been observed by Cf (III); therefore, motivation of this study is to determine the hydration bond distance of Es(III). Hydration structure of Es(III) was determined using synchrotron radiation based EXAFS analysis. Observed hydration bond distance of Es(III) was unexpectedly shorter than the expected from an extrapolation up to systematic actinide contraction from U(III) to Cf(III), we could first observe, namely "Einstein Break". It suggests that this breaking observation strongly could correlate with an electronic configuration in the inner core of Es(III) on the comparison with lanthanide series result, rather than hydration number change, according to the theoretical calculation and an estimation from atomic volume results.

1A07 Production of carrier-free RI tracer of alkali metal using bemsstrahlung

IKEDA, H.[1,2], KIKUNAGA, H.[2], WATABE, H.[1]
([1]Cyclotron and Radioisotope Center, Tohoku University, [2]Research Center for Electron Photon Science, Tohoku University)

[Introduction] Alkali metals are very important elements for plant research. Behavior of alkali metals in plants can be traced using carrier-free RI without affecting plant activity. We established a method for producing K tracer (K-42, 43) by irradiation with braking radiation and made it possible to investigate the dynamics of plants. Cesium (Cs), rubidium (Rb) and sodium (Na) tracers can also be produced by irradiation with bremsstrahlungs. In this study, we decided to individually examine the production amount and separation condition of carrier-free alkali metal RI.

[Experiment]The CaO, BaO or SrCO3 target was irradiated with bremsstrahlung radiation. Pure water and oxalic acid aqueous solution were added to the irradiated target to precipitate Ca, Ba or Sr as oxalate. The supernatant was charged to a column filled with cation exchange resin. After washing with 0.01 M HCl, the alkali metal RI was eluted with 1-1.5 M HCl.

[Results] Alkali metals coprecipitated when the target substance was precipitated were about 15-30%. Thus, the total chemical yields of Cs-136 and Rb-86 were about 60-80%. We drew the elution curves of Cs-136 and Rb-86. This result suggested that Cs-136 and Rb-86 could be supplied as multitracers at the same time.

1A08 Development and evaluate of the α-ToF detector for MRTOF-MS

NIWASE, T.[1,2], WADA, M.[3], SCHURY, P.[3], ITO, Y.[1,4], KAJI, D.[1], ROSENBUSCH, M.[1], KIMURA, S.[1], MORIMOTO, K.[1], HABA, H.[1], ISHIZAWA, S.[1,5], MORITA, K.[1,2], MIYATAKE, H.[3], WOLLNIK, H.[6]

([1]RIKEN Nishina Center, [2]Kyushu Univ., [3]KEK, [4]JAEA, [5]Yamagata Univ., [6]NMSU)

The atomic mass is a unique quantity for each nucleus. Precise mass measurement allows us to identify the atomic number as well as the mass number.

Recently, we succeeded in measuring the masses of fusion evaporation products provided from the GARIS-II with a multi-reflection time-of-flight (MRTOF) mass spectrograph. Next experimental plan is the measurement of the masses of superheavy nuclei (SHN) for identification of the mass number and atomic number. In the SHN region, the expected rate of reaction products is less than one event per day due to small reaction cross sections.

We developed an α-ToF detector to accurately distinguish a true event from large background events such as molecular ions. The time correlation between the time-of-flight signal and α decay signals can discriminate the background events. We have tested the α-ToF detector with an alpha source and confirmed that the correlation between the timing signal and the decay energy can be successfully measured using the α-ToF detector.

I will talk about the result of the α-ToF detector performance test, and also about the future experimental plan with our MRTOF devices for simultaneous mass and decay-property measurements.

1A10 Compton Camera(ETCC) imaging experiment using technetium isotopes.

HATSUKAWA,Y.[1], HASHIMOTO,K.[1], KABUKI, S.[2], KUSHIDA2, [J.], TSUKADA,K.[3,] ASAI, M.[3], TOYOSHIMA, A.[2], NISHIJIMA, K.[2], KUNIEDA, E[2].

([1]QST, [2]Tokai Univ., [3]JAEA)

Technetium-99m is used in radioactive medical diagnostic tests, for example as a radioactive tracer that a nuclear medicine gamma camera can detect in the human body. It is well suited to the role because it emits readily detectable 141 keV gamma-rays, and its half-life is 6.01 h (meaning that about 94 % of it decays to ^{99}Tc in 24 h).

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 require technetium isotopes emitting higher energy gamma-rays. A Tc isotope, ^{95}Tc ($T_{1/2} = 20$ h; $E\gamma = 765.8$ keV) is a candidate for tracer of the Compton camera imaging.

In this study, Technetium-95 was produced by the ^{nat}Mo(p,n)^{95}Tc reaction. The feasibility study of Compton camera imaging using gamma rays emitted from ^{95}Tc was carried out.

1A11 Calculation of Nuclear Reaction Cross Section in In-Flight Muon Catalyzed Fusion Cycle

KINO, Y. [1,2], YAMASHITA, T. [1], SATO, M. [2]

([1]Department of Chemistry, Tohoku University, [2]Faculty of Engineering, Chubu University[2])

The muon catalyzed fusion (μCF) has been expected to be a new energy source and mono-energetic neutron source. In the μCF, a muonic molecule formation is a rate determining process, and the obtained energy from the nuclear fusion reaction cannot exceed the energy for muon production. We newly propose an in-flight muon catalyzed fusion (IFμCF) cycle which avoids the rate determining process. In the IFμCF cycle, nuclear fusion reaction occurs during muon atomic collisions.

In this study, nuclear fusion reactions ($d\mu + t \rightarrow \alpha + n + \mu + 17.6$ MeV, d↔t) are calculated with a non-adiabatic coupled channel method [1]. The nuclear reaction cross sections are more than three orders of magnitude higher than typical cross sections used in the thermonuclear fusion study, but the temperature in the IFμCF is one order of magnitude lower than the thermonuclear fusion. Thus, if a high intensity muon beam is obtained, the IFμCF can be maintained by self-ignition.

[1] E. Hiyama, Y. Kino, M. Kamimura, Prog. Part. Nucl. Phys. 51, 223 (2003)

1A12 Development of a retarding-field magnetic-bottle electron spectrometer and measurements of the internal conversion electrons from U-235m (JNRS)

SHIGEKAWA, Y.[1], KASAMATSU, Y.[1], YAMAKITA, Y.[2], YASUDA, Y.[1], KONDO, N.[1], WATANABE, E.[1], SHINOHARA, A.[1]

([1]Graduate School of Science, Osaka University, [2]Graduate School of Informatics and Engineering, The University of Electro-Communications)

As ^{235m}U and ^{229m}Th have extremely low excitation energies, the nuclei interact with outer-shell electrons in the internal conversion (IC) process. Therefore, the half-lives of these nuclides vary depending on the chemical environments. We are aiming to elucidate the deexcitation process of ^{235m}U and ^{229m}Th by measuring the IC electron energy spectra in addition to the half-lives for various chemical environments. In order to precisely record the IC electron energy spectra of these nuclides, high detection efficiency as well as high energy resolution is required for the spectrometer. In this work, we designed and fabricated a retarding-field magnetic-bottle electron spectrometer. Moreover, we evaluated the collection efficiency of electrons to the channeltron detector under various magnetic fields. It was found that the collection efficiency of electrons becomes 100% on the appropriate magnetic condition. Next, the half-lives and the energy spectra of ^{235m}U were measured for ^{235m}U on the surface of Cu and for ^{235m}U that reacted with HF or HCl gas. With the high detection efficiency of this magnetic-bottle spectrometer, we were able to measure the half-lives and the energy spectra with high S/N and energy resolution. Especially, we clearly observed the difference of the half-lives and the energy spectra between ^{235m}U that reacted with HCl gas and ^{235m}U on the surface of Cu.

1B01 Existence forms of radiocesium in waters from river systems in Hama-Dori area in Fukushima Prefecture, Japan

NAGAO, S.[1], YAMAGISHI, E.[2], MOROKADO, T.[2], SHIMAMURA, A.[3], TOMIHARA, S.[3], OCHIAI, S.[1]

([1]INET, Kanazawa Univ., [2]College of Science and Engineering, Kanazawa Univ., [3]Aquamarine Fukushima)

Nuclear accident at the Fukushima Daiichi Nuclear Power Plant occurred after the 2011 Tohoku Earthquake and Tsunami. To estimate the impacts of radiation dose to human health, it is important to understand dynamics of radionuclides, especially ^{134}Cs and ^{137}Cs, in river watershed environments. The aim of this study is to investigate radioactivity of ^{134}Cs and ^{137}Cs in river systems in Fukushima Prefecture, and discuss with the transport of radiocesium from the surface soil to the river waters. The 20-100 L of river water samples were collected at monitoring stations in five river systems (Abukuma River, Niida River, Iwasawa River, Kido River and Natsui River) in Hama-Dori area from Fukushima Prefecture during 2015-2017. The ^{134}Cs and ^{137}Cs were separated by co-precipitation with AMP and measured by gamma-ray spectrometry using low BKG Ge detectors at the Low Level Radioactivity Laboratory, Kanazawa University. The particulate phase of ^{137}Cs ranged from 54% to 92% for the river waters from the Abukuma, Niida, Kido and Natsui River. On the other hand, the Iwasawa River has less than 10% for the waters with higher conductivity and DOC concentration. This indicates watershed condition is important to existence forms of radiocesium in the Fukuhsima river waters.

B02 Seasonal variation of the activity concentration of radioactive cesium in sediment sampled at Tamagawa River Watershed

AKIYAMA, M.[1], HAGIWARA, K.[2], KOIKE, Y.[2]

([1] Grad. Sch. of Sci. and Tech., Meiji Univ., [2] Sch. of Sci. and Tech., Meiji Univ.)

Monitoring of radioactive cesium is crucial for investigation of activity distribution and variation tracing of radioactive substances. In this study, radioactive cesium in sediments sampled at 45 points in Tamagawa River watershed, concluding main and tributary stream, were determined for behavior analysis of radioactive cesium at low dose areas. In the mainstream at Tamagawa River watershed, the activity concentrations of radioactive cesium in sediment sampled at downstream were higher than that at upstream and midstream. On the other hand, the concentrations of radioactive cesium in sediment sampled at Nogawa river, Hirase river, and Nikaryo carnal were high compared to the mainstream. The flow rate of the tributaries at Tamagawa River Watershed is slower than that of the mainstream. These results indicated that the activity concentration of ^{137}Cs in sediment increased by locally accumulating ^{137}Cs in slowly flowing river water.

1B03 Contribution of groundwater discharge to bottom water of Nanao West Bay based on ^{222}Rn horizontal distribution.

FUJITA, A.[1], NAGAO, S.[2], OCHIAI, S.[2], SUGIMOTO, R.[3],MATSUNAKA, T.[2], M. Charette[4], P. Henderson [4]

([1]Graduate School of Natural Science and Technology, Kanazawa Univ., [2]Institute of Nature and Environmental Technology, Kanazawa Univ., [3]Reseach Center for Marine Bioresources, Fukui Prefectural Univ., [4]Department of Marine Chemistry and Geochemistry, Woods Hole Oceanographic Institute)

Biological production is more active in coastal areas than open sea because large amount of nutrients from terrestrial areas are carried by rivers. On the other hand, contribution of nutrients from groundwater to coastal areas is also important, but identification of submarine groundwater discharge (SGD) is tedious work. Many scientists have been using ^{222}Rn to specify the contribution of groundwater. ^{222}Rn is a daughter nuclide of ^{226}Ra, water-soluble and noble gas. The sedimentary layer is rich in ^{238}U and ^{226}Ra which belong among Uranium series, so the concentration of ^{222}Rn is higher in groundwater than surface water. Moreover ^{222}Rn has a half - life of 3.83 days and is used to specify the contribution of groundwater with short residence time. In this study, research was carried out at Nanao West Bay in October of 2017 and April and July of 2018. In October of 2017, the higher concentration is observed at the center-east area in the bay. But the higher concentration site is detected at the southwest area in April of 2018 and are the northeast and southwest areas in July of 2018. It is necessary to continue the survey to grasp the actual condition of SGD through the year.

1B04 Evaluation of Analytical Method for ^{210}Po/^{210}Pb in Marine Product using Interlaboratory Comparison

MIURA, T.[1], OTA, T.[2], TERADA, A.[3]

([1]National Metrology Institute of Japan, [2]Japan Chemical Analysis Center, [3]National Institute of Public Health)

Marine products such as marine fish are well known to contain ^{210}Po at relatively high concentrations. It is important to precisely measure ^{210}Pb which is a substantial parent nuclide of ^{210}Po and ^{210}Po in marine products from the viewpoint of internal exposure dose evaluation attributable to food. We verified the analytical method applied to the measurement of ^{210}Po/^{210} Po in food samples using a freeze-dried powder of fish meat (flounder) 5 years after collection and a ^{210}Po solution and a ^{209}Po standard solution separated from a commercially available metallic lead reagent. The loss of Po for each chemical separation step (acid decomposition, extraction chromatography, measurement source preparation by electrodeposition) of the analysis method was examined. As a result, there was no significant loss of Po in the chemical separation operation. Therefore, as a next step, we tried to evaluate the performance of the ^{210}Po/^{210}Po analysis method by conducting a interlaboratory comparison in which participate several laboratories analyze test samples based on the standard analysis procedure document.

1B05 Elucidation for production scenario of insoluble Cs particles from Unit 1 using multifaceted analysis

SATOU, Y.[1], KOBATA, M.[1], OKANE, T.[1], SUEKI, K.[2], MATSUO, K.[2],ISHIII, K.[2], Martin, P. G.[3], Scott, T. B.[3]

([1]JAEA, [2]Graduate School of Pure and Applied Science, Univ. of Tsukuba, [3]Interface Analysis Centre, Univ. of Bristol)

In order to disclose production process of insoluble Cs particles ejected from the Unit 1 of Fukushima Dai-ichi Nuclear Power Station ("Type B" particles), we employed laboratory X-ray computed tomography (Lab. CT), synchrotron-μ-X-ray tomography (XRT), and Hard X-ray Photoelectron Spectroscopy (HAXPES). Type B particles were isolated from ground dust sample obtained from 3km north from the Fukushima Daiichi Power Station. Type B contain bubbles in the body and detected fragments of heavier material – steel, caesium, fuel. These elements are spread outer side, also high-absorption area of X-ray is surrounding of the particle. Some elements such as Fe showed metallic and low oxide state using a HAXPES. Elements except silicon in Type B were expected that impurities of source material. However, above results shows the elements were attached on the particle during solidification process.

1B06 Quantity of trace element in insoluble particle including radioactive Cs found in Okuma town, Fukushima prefecture by destruction analysis

IGARASHI, J.[1], ZHANG, Z.[1], NINOMIYA, K.[1], SHINOHARA, A.[1], SUEKI, K.[2], SATOU, Y.[3]

([1]Graduate School of Science, Osaka University., [2]Graduate School of Pure and Applied Sciences, Tsukuba University., [3]JAEA.)

Insoluble radioactive Cs particles(insoluble particles) made of mainly SiO_2 were released by the accident of Fukushima Dai-ichi Nuclear Power Plant. It was considered that the insoluble particles were formed in the reactor during the accident. In the previous studies, the insoluble particles can be classified to at least two types from the properties and radioactivity concentration of Cs. In this study, the insoluble particle which was approximately 100 μm in diameter and low radioactivity concentration of Cs was found in Okuma town, Fukushima prefecture. It was estimated that this insoluble particle was derived from unit 2 or 3 reactor by its radioactive ratio, ^{134}Cs/^{137}Cs.To investigate the formation process of this insoluble particle in the reactor, we focused on the amount of the non-volatile element, Sr in the insoluble particle. In this study, we dissolved the insoluble particle by the alkali fusion method and extracted Sr by the Sr-Rad Disk. Finally, we quantified the amount of the Sr by measuring the ^{90}Sr using the Cherenkov light measurement. As a result of the analysis, it was found that the radioactive ^{90}Sr of the insoluble particle was approximately 0.04 Bq.

1B07 Development on the Determination Method of Radioactive Antimony and the Obtained Distribution of Its Soil Deposition near Fukushima Daiichi Nuclear Power Plant

MIYAZAWA, N.[1], UESUGI, M.[2], YOKOYAMA, A.[3],

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

A large amount of radionuclides were released into the atmosphere and deposited on soil due to the accident of Fukushima Daiichi Nuclear Power Plant (FDNPP) in 2011. Lots of studies focused on isotopes of cesium and iodine. However, behaviors for the other radioactive elements are not well unknown. For such study on a nuclide of interest, we have researched for ^{125}Sb ($T_{1/2} = 2.75$ y). Antimony, a volatile element like cesium, is seldom reported, because the deposited amount of ^{125}Sb, its long-lived isotope, is relatively small and counting photons in peak area is interfered with γ-rays from abundant radioactive cesium. In our previous study, we established a determination method of ^{125}Sb with a removal process of cesium by using ammonium phosphomolybdate adsorption method or an anion exchange resin method. In this work, we obtained the analytical results under some leaching conditions for antimony in soil, and showed the distribution of ^{125}Sb near the FDNPP.

1B08 Atmospheric Radiocesium Concentrations in East Japan just after TEPCO FDNPP Accident by Analysis of Tape Filters Used at Automatic SPM Monitors.

OURA, Y.[1], TSURUTA, H.[2], EBIHARA, M.[3], MORIGUCHI, Y.[4], OHARA, T.[5], NAKAJIMA., T.[6]([1]Tokyo Metropolitan Univ., [2]RESTEC, [3]Waseda Univ., [4]Univ. of Tokyo, [5]NIES, [6]JAXA)

We have been determining ^{134}Cs and ^{137,}Cs in suspending particulate matters (SPM; aerosols less than 10 μm in diameter) collected hourly from Mar. 12 to 23, 2011 at SPM monitoring stations in eastern Japan to reveal spatio-temporal trajectory of radioactive plumes caused by TEPCO FDNPP accident, and the dataset for 99 monitoring stations was reported. After that, radiocesium concentrations at additional 48 stations from Aomori prefecture to Shizuoka prefecture were determined. In Aomori prefecture, no radiocesium was detected from Mar.12 to 25. In Iwate prefecture ^{137}Cs with max. about 4 Bq/m^3 was detected at some stations on Mar.13 and 21. In Nagano prefecture, radioactive plume with ^{137}Cs of max. about 30 Bq/m^3 was observed on Mar.15 at west side of Kanto-Santi but no plume was observed at northern area on same period. In shizuoka prefecture, radioactive plume with ^{137}Cs of max. 0.8 Bq/m^3 was observed at western area on Mar. 21 but was not at eastern area. It seems that the plume went through on the ocean and reach at western area.

1B09 Transfer behavior of Sr-90 from environment to cattle teeth after the Fukushima Daiichi Power Plant accident.

KOARAI, K.[1], KINO, Y.[1], NISHIYAMA, J.[1], KANEKO, H.[1], ONO, T.[1], OKA, T.[1,2], TAKAHASHI, A.[3], SUZUKI, T.[4,5], SHIMIZU, Y.[4], CHIBA, M.[4], OSAKA, K.[4,5], SASAKI, K.[4], URUSHIHARA, Y.[6], FUKUDA, T.[7], ISOGAI, E.[8], SEKINE, T.[1,2], FUKUMOTO, M[9]. and SHINODA, H[4].

([1]Department of Chemistry, Tohoku University, [2]IEHE, Tohoku University, [3]Tohoku University Hospital, [4]Graduate School of Dentistry, Tohoku University, [5]IRIDeS, Tohoku University, [6]Tohoku University School of Medicine, [7]Faculty of Science and Engineering, Iwate University, [8]Graduate School of Agricultural Science, Tohoku University, [9]Tokyo Medical University)

We have investigated transfer behavior of Sr-90 from soil to cattle teeth after the Fukushima Daiichi Power Plant (FNPP) accident. The cattle and soil were collected in Okuma town, which was in the ex-evacuation area of the FNPP accident. The soluble fractions of the soil were extracted by milli-Q water or 1M CH_3COONH_4 solution. Radio activity of Sr-90 in the samples was determined after the chemical separation (fuming nitric acid method or Sr resin method). Stable Sr and Ca in the samples were determined by ICP-MS or ICP-AES. Solubility of Sr-90 was larger than those of stable Sr and Ca. The result implies that the nuclide could easily migrate from the soil to the teeth. The specific activity of the soluble fraction was similar to that of the teeth developed after the FNPP accident. The result indicates that the soluble Sr-90 in the soil would be the source of the Sr-90 in teeth. The ratio of Sr-90 to Ca of the soluble fraction was higher than that of the tooth. This indicates that Sr-90 was discriminated from Ca in the incorporation process.

1B10 Systematics of adsorption structure of various hard metal ions on phyllosilicates: comparison of strontium and cesium

YAMAGUCHI, A.、KURIHARA, Y.

(Graduate School of Science, The Univ. of Tokyo)

Extend X-ray adsorption fine structure (EXAFS) spectra of some alkaline ions, alkaline earth ions, and rare earth ions adsorbed on clay minerals were obtained in this study. Theses systematic results revealed that ions larger than barium (Ba^{2+}) forms inner-sphere (IS) complex and smaller outer-sphere (OS) complex, during the adsorption on clay minerals. This implication is helpful to explain and predict the behavior of various ions in environment. This study aimed to reveal local structures of strontium (Sr) and cesium (Cs) adsorbed on clay minerals (vermiculite and montmorillonite) and understand stabilities of the adsorbed species of ^{90}Sr and ^{137}Cs on the clay minerals in the environment. In this study, adsorption experiments for determination of solid-water distribution, XRD for interlayer distances of Sr or Cs adsorbed clay minerals, and

EXAFS for speciation of the adsorbed Sr and Cs were conducted. They showed that Sr and Cs formed outer and inner-sphere complexes, respectively, in the interlayer, which explains the different degree of mobility of ^{90}Sr and ^{137}Cs in the environment.

1B11 Fixation of Ra by specific adsorption on clay minerals based on analyses of core samples in Ningyo-toge uranium deposit

YAMAGUCHI, A. [1], KURIHARA, Y. [2], TANAKA, K. [3], OHARA, Y. [2], KOZAI, N. [3], TAKAHASHI, Y. [1]

([1]Graduate School of Science, The Univ. of Tokyo, [2]JAEA NEEC, [3]JAEA ASRC)

Our previous study suggested that cations with large ionic sizes have strong affinity as adsorbed species into the interlayer of clay minerals, which suggested that radium (Ra^{2+}) is strongly adsorbed on clay minerals. However, actual adsorption behavior of Ra^{2+} in the environment has not been well understood. Therefore, the purpose of this study is to clarify the role of clay minerals in the migration of Ra^{2+} in geosphere using core samples collected in Ningyo-toge uranium deposits. Ningyo-toge is an appropriate site for this purpose, because (i) the site contains large amounts of uranium (U) and Ra and (ii) weathered granite layer is found under the uranium ore-bearing layer. These factors enable us to study the migration of Ra^{2+} in terms of fixation of Ra^{2+} by clay minerals in natural environment. Activity ratio of $^{226}Ra/^{238}U$ measured by gamma-ray spectrometry showed that Ra^{2+} was selectivity fixed in the weathered granite layer. Furthermore, XAFS analyses of iron (Fe), manganese (Mn), and barium (Ba) suggested that the host phase of Ba^{2+} is clay minerals rather than Fe-Mn oxides and carbonate, which is likely the case for Ra^{2+}. These results indicated that Ra^{2+} leached into groundwater from U concentrated layer was selectivity fixed on clay minerals, which supports the effectiveness to estimate adsorption behavior of various metal ions depending on the ionic radius.

1B12 Application of simple and rapid radiostrontium analysis to natural seawater

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

([1]Radioisotope Res. Facility, Jikei Univ., [2]Healthcare BU, Hitachi Ltd., [3]Aichi Medical Univ., [3]RIC Medical Division, Nagoya Univ.)

A safe, simple and rapid analysis method of radiostrontium in environmental sample was developed and was applied to natural seawater. Surface seawater at the coast of Fukushima and Mie prefecture was examined. One litter of seawater was passed through a column (CV=60 mL) with a cation exchange resin (Dowex 50W-X8). Ammonium acetate-methanol solution was flowed through the column to eluate Ca, etc., and then Sr was eluted with 4M-HCl. Sr was collected as carbonate precipitate on a membrane filter. Immediately after the chemical separation, the β-rays were measured by a low background liquid scintillation system (AccuFLEX LSC-LB7, Hitachi, Ltd.) using a plastic scintillator bottle. The same sample was intermittently measured for the growth of ^{90}Y until radiation equilibrium (about 3 weeks later). ^{90}Sr concentration was determined with Sr yield of 70% based on the prior tracer experiments. The ^{90}Sr concentrations in this work were all below the minimum detectable concentration（MDC）. The ^{90}Sr MDC of these samples were 0.02-0.6 Bq L^{-1}. It took 2 days (operation time about 10 hours) for chemical separation. It became possible to analyze more quickly than the conventional method (20 days). This is a useful method that can be used for screening of radiostrontium in seawater.

1B13 Spectrum analysis of plastic scintillator bottle via liquid scintillation counter

KATO, Y.[1], MINOWA, H.[2], KOJIMA, S.[3], OGATA, Y.[4]
([1]Healthcare BU, Hitachi Ltd., [2]Radioisotope Res. Facility, Jikei Univ., [3]Aichi Medical Univ., [4]RIC Medical Division, Nagoya Univ.)

Conventional methods of radiostrontium analysis are complex procedures, require time for chemical separation together with large amounts of deleterious substances. Safe, simple and rapid analytical method is desired. We tried to develop a new method to analyze radiostrontium concentration in seawater. A measurement method using a plastic scintillator bottle (PSB) is reported, especially the spectral analysis. Filter samples obtained by a chemical separation (see the former report) were measured using PSBs via a low background liquid scintillation system, LSC-LB 7 (Hitachi Ltd.). The minimum detectable concentration was 0.02 Bq L^{-1}, which was obtained with 1-L sample, background 0.14 cps, counting efficiency of 70%, and chemical yield 70%. It was 1/1500 of the legal concentration limit of ^{90}Sr in the wastewater. Therefore this method is proved to be effective as a screening method. In addition, from the spectrum analysis of ^{90}Sr - ^{90}Y to be measured and ^{210}Pb considered to be an interfering substance, the possibility of nuclide identification by using the PSB was suggested.

2A01 Development of non-destructive isotope analysis method using negative muon

KUDO, T.[1], NINOMIYA, K.[1], KUBO, K.[2], SHINOHARA, A.[1], TAMPO, M.[3], MIYAKE, Y.[3]
([1]Graduate School of Science, Osaka Univ., [2]College of Liberal Arts, International Christian Univ., [3]Institute of Materials Structure Science, High Energy Accelerator Research Organization)

When a negative muon stops in a material, a muonic atom that has the muon in the atomic system instead of an electron is formed. The captured muon releases muonic X-ray by muon de-excitation process. Due to a large mass of a muon, muonic X-ray has high energies and very hard. Using these properties, development of non-destructive elemental analysis by muon has been on going.

In this study, we focused on the isotopic shift of muonic X-ray for developing non-destructive isotope analysis method. The muon irradiation experiment for ^{nat}Pb and enriched ^{208}Pb metal samples was carried out at D2 beam line in J-PARC/MLF, and emitted muonic X-rays were measured by five Germanium detectors.

From muonic X-ray spectra, we observed isotopic shift of muonic Pb KX-ray and determined muonic X-ray intensity of ^{208}Pb, ^{207}Pb and ^{206}Pb, respectively. From muonic X-ray intensity, we determined isotopic abundance of ^{208}Pb as 51.5±5.8%, ^{207}Pb as 24.5±5.4% and ^{206}Pb as 22.2±4.8% for ^{nat}Pb, these values are consistent with the reported isotopic abundance.

2A03 Composition of Fe-bearing minerals in Antarctic meteorites MIL 07710 and Y-790272

SATO, W.[1,2], NAKAGAWA, M.[2], SHIRAI, N.[3], EBIHARA, M.[4]

([1]Institute of Science and Engineering, Kanazawa Univ., [2]Graduate School of Natural Science and Technology, Kanazawa Univ., [3]Graduate School of Science and Engineering, Tokyo Metropolitan Univ., [4]Faculty of Education and Integrated Arts and Sciences, Waseda Univ.)

^{57}Fe Mössbauer spectroscopy was applied to compositional studies of Fe-bearing minerals in two different ordinary chondrites, Miller Range 07710 (MIL 07710) and Yamato 790272 (Y-790272), collected in Antarctica. For both samples, spectral analysis revealed that there are five quadrupole doublets and two magnetic sextets in the room-temperature spectra, suggesting the presence of olivine (M1 and M2), pyroxene (M1 and M2), a high-spin Fe^{3+} containing phase, antiferromagnetic FeS (troilite), and ferromagnetic Fe-Ni alloy. The large relative area of the oxidized Fe^{3+} phases and the small intensity of the metal phases signify a possible effect of terrestrial weathering especially for Y-790272 (H4). In the session, we discuss the unknown Fe^{3+} phases based on low-temperature Mössbauer spectra.

2A05 Structure of Solubility-limiting Solid Phase in M(IV) Hydroxide System using X-ray Spectroscopy

KOBAYASHI, T.[1], NAKAJIMA, S.[1], MOTOKAWA, R., MATSUMURA, D., SAITO, T., SASAKI, T.[1]

([1]Graduate School of Engineering, Kyoto Univ., [2]Japan Atomic Energy Agency, [3]School of Engineering, The University of Tokyo)

The structure of zirconium(IV) hydroxide solid phase after aging at elevated temperatures was investigated by extended X-ray absorption fine structure (EXAFS), wide- and small-angle X-ray scattering (WAXS/SAXS) to elucidate a process on the transformation of solubility-limiting solid phase. Sample solutions with amorphous Zr hydroxide ($Zr(OH)_4(am)$) were prepared at certain pH and stored at 25, 40, 60 and 90°C. Aliquots of the solid phases after aging were then examined by Zr K-edge (18.00 keV) EXAFS at BL14B1 of SPring-8, and WAXS/SAXS at BL8S3 of Aichi Synchrotron Radiation Center. The SAXS profiles after aging at 25°C to 60°C suggested that the solid phases were comprised of primary particles having a size of approximately 3 nm and their aggregates. After aging at 90°C, the solid phase consisted of those with the size larger than 30 nm in the acidic pH and the size altered depending on pH. The WAXS spectra indicated that the crystallization occurred after aging at 90°C. In the EXAFS, all spectra exhibited intense Fourier-transfomred peaks at ~1.7 Å and 3.0 Å, representing O and Zr atoms in the coordination sphere. The intensities of these peaks suggested that the primary particles after aging at 25°C to 60°C consisted of tetrameric hydrolysis species and a further polymerization proceeded at 90°C.

2A07 Synthesis, structure, and electronic configuration of the luminescent nitridotechnetium complex with a tridentate ligand coordination site

YOSHIMURA, T.[1], NAGATA, K.[1], SHIROYAMA, T.[2], KINO, Y.[3], TAKAYAMA, T.[4], SEKINE, T.[5], SHINOHARA, A.[1,2]
（[1]Institute for Radiation Sciences, Osaka Univ., [2]Graduate School of Science, Osaka Univ., [3]Graduate School of Science, Tohoku Univ., [4]Daido Univ., [5]Institute for Excellence in Higher Education, Tohoku Univ.）

Technetium has been widely used as a radiopharmaceutical for diagnosis. Recently, an interest is growing in a molecule having bimodal imaging ability by photoemission and radiation from a nucleus. In this study, we synthesized a *cis*-dicyanonitridotechnetium(V) core which is small in size, has low molecular weight, and is luminescent. Moreover, the core has the sites to which a tridentate ligand coordinates. Treatment of an orange solution of $(n\text{-}C_4H_9)_4N[^{99}TcNCl_4]$ with 3 equiv. bpa gave $[TcNCl_2(bpa)]$ (**1**). The reaction of **1** with 2 equiv. CN^- yielded $[TcN(CN)_2(bpa)]$ (**2**). The shift of the d-d transition energies in UV-vis spectra would be in accordance with the number and nature of the equatorial ligands; the ligand strength of a spectrochemical series (Cl^- < pyridine < CN^-) affects to the d-d transition energy, and increasing the number of the ligands contributing to a larger ligand field splitting parameter increases the electronic transition energy in the d^2 nitrido Tc complexes. Photoemission with a maximum wavelength at 666 nm was observed for **2** in the solid-state at 298 K. The trend of the emission energies suggests that the ligand strength of the spectrochemical series for the equatorial ligands is important for the emission energy.

2B01 Measurement of high-energy prompt γ-rays from neutron induced fission of ^{235}U

MAKII, H.[1], NISHIO, K.[1], HIROSE, K.[1], ORLANDI, R.[1], LÉGUILLON, R.[1], OGAWA, T.[1], SOLDNER, T.[2], HAMBSH, F.-J.[3], AÏCHE, M.[4], ASTIER, A.[5], CZAJKOWSKI, S.[4], FROST, R.[6], GUO, S.[4], KÖSTER, U.[2], MATHIER, L.[4], OHTSUKI, T.[7], PETRACHE, C.M.[5], POLLITT, A.[2], SEKIMOTO, S.[7], TAKAMIYA, K.[7], TSEKHANOVICH, I.[5]
([1]JAEA, [2]ILL, [3]JRC, [4]CENBG, [5]CSNSM, [6]Univ. of Manchester, [7]KURNS)

The Japanese government has a plan to remove the fuel debris for the full deconstruction of Fukushima I Nuclear Power Plants. The fuel debris must be removed without reaching criticality. The criticality of the system must be monitored by detecting the fission rate in the debris. The fission rate should be measured by detecting radioactivity from fission, which has different characteristics from background γ rays coming from long-lived fission products. One plausible idea is to measure the high-energy prompt fission γ rays, whose energies are high enough to be separated from the background γ rays. However, the measured γ-ray spectra for neutron induced fission of ^{235}U are limited below the γ-ray energy of about 7 MeV. This prompted us to make a new measurement to extend γ-ray spectrum limit to higher energies. The measurement has been carried out at the PF1b cold-neutron beam facility of the Institut Laue-Langevin (ILL), Grenoble, France. We have successfully measured the γ-ray spectrum up to energies of about 20 MeV. In this contribution we will present the results obtained the measurement.

2B02 Simulation of bremsstrahlung gamma ray from winter thunderclouds which were tilted along the slope of low mountains

KUROSAKI, H.

(Niigata Prefectural Institute of Environmental Radiation Monitoring)

Several detection of bremsstrahlung gamma ray from winter thunderclouds along the coast of the Sea of Japan had been reported. We observed bremsstrahlung gamma rays four times by using the NaI(Tl) scintillator at an altitude of 207 m, about 8 km inland from the coast, between from Jan. to Mar. 2018., under the weather conditions that wind directions were south ~ southwest and the wind speed of 3.3 ~ 6.7 m / s were blowing. However, these were different from the condition that thunderclouds were tilted by strong winter storm along the coast, such as the condition of use of the model that we have named“Tilted searchlight radiation model”, which was invented in 2011. But, results of count rate calculated from this model were in good agreement with measurement data. Accordingly, we thought that bremsstrahlung gamma ray emissions from the tilted thundercloud were occurred. Since the thunderclouds height of 313 ~ 371 m and the low mountain height of 351 m was almost same, the thunderclouds bumped into the mountain and tilted along the slope of the mountain by the weak wind of southerly. It was first observation of gamma ray emission from thunderclouds which were tilted along the slope of low mountains.

2B03 Reduction of radiocesium concentration from soil and ash using chlorination-vaporization process

KINOSHITA, N.[1], JIAO, F.[2], ASADA, M.[1], KAWAGUCHI, M.[1], HONDA, M.[3], SUEKI, K.[3], NINOMIYA, Y.[2]

([1]Shimizu Co., [2]Chubu Univ., [3]Univ. Tsukuba)

The reduction of amount of waste discharged from decontamination work is one of the important issues. We applied the chlorination-vaporization process to soil samples collected in Fukushima prefecture and clinker obtained by burning rice straw. These samples mixed with known amount of $CaCl_2$ or NaCl were heated at 900 - 1200 °C using an electric furnace for 30 min. The vaporization ratio and feature of radiocesium distribution were measured with gamma-ray spectrometry using a HPGe and imaging plate, respectively. Significant difference was not observed in the vaporization ratio from samples mixed with NaCl+CaO and $CaCl_2$+CaO. The vaporization ratio of radiocesium reached up to 95% when the samples were heated at higher than 1100 °C. On the other hand, hot particles were observed in the soils. When the soil heated at 900 °C, radiocesium showed homogeneous distribution once. Then, the cesium vaporized.

2B04 A new technique for removing radioactive nuclides from aqueous solution by coprecipitation with barite

TOKUNAGA, K., KOZAI, N., and TAKAHASHI, Y.

([1]Ningyo-, [2]ASRC-JAEA, [3]Univ. of Tokyo)

In the present study, we explore a new application of barite (BaSO4) as a sequestering phase for selenite (Se(IV)) and selenate (Se(VI)) ions from aqueous solutions. Barite is a common phase in many geological environments, and it can be used to remove toxic and/or radioactive elements from aqueous solutions, but it has not been widely used in environmental studies. This study describes the mechanisms of Se distribution between barite and water, thus providing a good estimate of its ability to effectively remove Se(IV) and Se(VI) from aqueous solutions (more than 80%) using the optimized experimental parameters examined here.

2B05 ^{129}I/^{131}I ratios for the FDNPP-derived radioactive aerosol materials transported to the Tokyo metropolitan area

EBIHAEA, M.[1], OURA, Y.[2], SHIRAI, N.[2], NAGAKAWA, Y.[3], SAKURAI, N.[3], HABA, H.[4], MATSUZAKI, H.[5], TSURUTA, H.[6], MORIGUCHI, Y.[7]

([1]Waseda Univ., [2]Tokyo Metropolitan Univ., [3]Tokyo Metro. Indus. Tech. Res. Inst., [4]RIKEN, [5]Univ. Museum, Univ. of Tokyo, [6]RESTEC, [7]Grad. School of Eng., Univ. Tokyo)

A large amount of radioactive materials were dispersed in the environment by the FDNPP accident in 2011. To retrieve the diffusion trajectory of ^{131}I, airborne particulate matters (APMs) collected in the Tokyo metropolitan area just after the accident and measured for their ^{131}I activity were analyzed for their ^{129}I contents by AMS. In evaluating blank levels of chemicals and filters used for collecting APMs, we established the analytical procedure for determining ^{129}I activity of as low as 10^{-8} Bq for a small piece of filter samples (about 0.1 cm^2). Coupled with ^{131}I data, activity ratios of ^{129}I/^{131}I were obtained for 43 APM samples and a mean value of 2.18×10^{-8} (±34% for 1s) was obtained. This value is smaller than a mean value of soil samples (by 25-30%) and the inventory data (by 40%). Such a difference can be explained in terms of desorption loss of ^{129}I from APMs. The same procedure was applied to suspended particulate matter (SPM) samples collected at an SPM monitoring station located at the nearby APM-collecting site, and consistent results with those of APMs in terms of ^{129}I concentration were obtained. The new approach developed in this study for estimating ^{131}I contents in particulates by measuring ^{129}I in APM/SPM samples can be the first step for evaluating the internal radiation exposure by inhalation of ^{131}I.

2B06 Study on ocean circulation in the Japan Sea using iodine-129

MATSUNAKA, T.[1], NAGAO, S.[1], INOUE, M.[1], OCHIAI, S.[1], SASA, K.[2], MORITA, T.[3], MIKI, S.[3], HONDA, N.[3], KUDO, I.[4], TAKIKAWA, T.[5], ARAMAKI, T.[6], HONDA, M.[2], SUEKI, K.[2]
([1]Kanazawa Univ., [2]Univ. of Tsukuba, [3]Japan Fisheries Research and Education Agency, [4]Hokkaido Univ., [5]Nagasaki Univ., [6]National Institute for Environmental Studies)

Oceanographic observation revealed that warming and oxygen decrease of the Japan Sea Bottom Water (JSBW) are responding to air temperature raise in winter. The investigation of water dynamics in the Japan Sea using radioactive tracer is essential for elucidating the interaction between climate change and the convection systems. Anthropogenic ^{129}I ($T_{1/2}$ = 15.7 million years) produced from the thermal neutron fission, is dominated by release from nuclear fuel reprocessing plant. To illuminate the availability of ^{129}I as a tracer of surface circulations and vertical convections in the Japan Sea, we investigated the horizontal and vertical distributions of the ^{129}I at the large area in 2017–2018. The dissolved ^{129}I in surface water varied from 17.8 to 23.8 nBq L^{-1} at the area of 38–46°N and 135–141°E, and was negatively correlated with salinity ($R^2 = 0.82$). This distribution revealed that the dissolved ^{129}I in the area was controlled by mixing of water mass from the Liman Current and the Tsushima Current. Meanwhile, the dissolved ^{129}I in the JSBW of the Japan Basin in 2017 was 4.2 nBq L^{-1}, which increased by 1.2 nBq L^{-1} in comparison to that in 2007. The ^{129}I is considered to be a tracer for the surface subarctic front and JSBW in the Japan Sea.

2B07 Searching for the origin of the Santan trade from radiocarbon dating and historical study

ODA, H.[1], NAKAMURA, K.[2]
([1]ISEE, Nagoya Univ., [2]National Institute of Technology, Hakodate College)

In Japan, silk fabrics called Ezo-nishiki are distributed throughout Hokkaido and northeastern Honshu. They were originally made in China, but they were also given to the chieftains of tribes and villages in the Amur Basin and imported to Sakhalin Island over the Mamiya Strait. They were then spread southward into Hokkaido by Ainu people living on Sakhalin Island and Hokkaido. The trade called the Santan trade peaked in the latter half of the 18th century to the early 19th century, but the origin was not clarified. Ezo-nishiki was the main merchandise of the Santan trade. However, there is no remaining Ezo-nishiki confirmed to have been made before the 18th century. We thus applied radiocarbon dating to Ezo-nishiki samples to determine their age and to clarify the origin of the Santan trade. We analyzed 34 samples of Ezo-nishiki by accelerator mass spectrometry. Calibrated radiocarbon ages of 33 of the samples indicated fabrication since the middle 17th century, confirming that the Santan trade peaked during the Qing Dynasty (1644–1912). One sample indicated fabrication between the first half of the 14th and the early 15th century. Ezo-nishiki thus provides evidence showing that the Santan trade originated at least as early as that time.

3A01 Development of dry-distillation method of ^{211}At for targeted alpha therapy

TOYOSHIMA, A.[1], ZHANG, Z.[2], KANDA, A.[2], IKEDA, T.[2], ICHIMURA, S.[2], OOE, K.[3], NAGATA K.[1,4], YOSHIMURA, T.[1,4], SHINOHARA, A.[1,2]

([1]Institute for Radiation Sciences, [2]Graduate School of Science, Osaka Univ., [3]Graduate School of Medicine, Osaka Univ., Radioisotope Research Center, Osaka Univ.)

In recent decades, targeted alpha therapy (TAT) has an increasing attention. TAT is expected to enable the effective cancer treatment by accumulating α-emitter-labelled compounds in tumor. At Osaka University, we are conducting the TAT project with ^{211}At in the collaboration among Research Center of Nuclear Physics (RCNP), Graduate School of Science, and Graduate School of Medicine. In this project, we need to prepare purified ^{211}At solution which is of sufficiently high quality to perform pharmaceutical developments and animal tests with ^{211}At. In this study, we examined separation conditions using our developed dry-distillation apparatus of ^{211}At. Astatine-211 was produced in the ^{209}Bi(α, 2n)^{211}At reaction at RCNP. After the irradiation, dry distillation was carried out. In a typical procedure, He and O_2 mixed gas was used and a small amount of water was included in the system. The Bi target was heated at 840^{o}C. Vapored ^{211}At was transferred to the cyrotrap through Teflon capirally. After 30-45 min, trapped ^{211}At was stripped with 100 μL of distilled water. The effluent was collected in a small vial and then supplied to the following experiments. At present, chemical yield of ^{211}At was 80 - 90% under optimum conditions. In the symposium, details of separation experiments will be presented.

3A02 The difference of ^{211}At species obtained by dry distillation depends on the elapsed time after separation

IKEDA, T.[1], TOYOSHIMA, A.[2], KANDA, A.[1], ICHIMURA, S.[1], ZHANG, Z.[1], YOSHIMURA, T.[1,3], NAGATA, K.[1,3], OOE, K.[4], SHINOHARA, A.[1,2]

（[1]Graduate School of science, Osaka Univ., [2]Institute for Radiation sciences, Osaka Univ., [3]Radioisotope Research Center, Osaka Univ., [4]Graduate School of medicine, Osaka Univ.）

Astatine-211 (^{211}At) attracts a lot of attention as an alpha-emitting radionuclide suitable for targeted alpha therapy. In purification of ^{211}At, a dry distillation method is often utilized. However, little is known about chemical species of ^{211}At in finally obtained aqueous solution after the distillation. Thus, we studied extraction and thin-layer chromatography (TLC) behavior of ^{211}At obtained by the dry distillation and also investigated the dependence of species ratios on the elapsed time after separation. After the irradiation of α beam on a ^{209}Bi target, ^{211}At was purified by the dry distillation. In the extraction experiment, after shaking for 2 min – 2 h, 350 µL of both of its phases (hexane and 1 M $HClO_4$) were precisely taken and the distribution ratio D was evaluated. As a result, some of the ^{211}At in distilled water exists as a neutral species which is extractable by hexane. The TLC analysis was also performed for the ^{211}At samples in distilled water. In the experiments, different 3 spots of ^{211}At with R_f values around 0, 0.5 and 0.9 were observed. It was also found that the percentage of cationic species with $R_f = 0$ increased as an increase of the elapsed time after separation. These results suggest that the relatively slow reaction of ^{211}At proceeds in aqueous solution just after dissolution.

3A03 Evaluation of At-211 Labeled Amino Acid Derivative for Targeted Alpha Therapy

ZHANG, Z.[1], KANEDA. K.[1], TOYOSHIMA, A.[2], SHINOHARA, A.[1], FUKASE, K.[1,2]

([1]Graduate School of Science, Osaka Univ, [2]Institute for Radiation Science, Osaka Univ)

Targeted alpha therapy (TAT) is an approach to selective delivery of an alpha emitter to tumor for killing cancer cells with its high linear energy transfer (LET) alpha-particle, while minimizing damage to surrounding cells. Astatine-211 has been recognized one of the most promising radionuclides for the TAT. Our group has developed L-[^{211}At]-α-Methyltyrosine (^{211}At-AAMT). The ^{211}At-AAMT is an analog compound of ^{18}F-FAMT which is an amino acid tracer for PET imaging and is accumulated in tumor cells via the L-type amino-acid transporter 1 (LAT1).

The ^{211}At-AAMT was synthesized in one-step reaction from L-α-methyltyrosine via mercuration, and then purified with ion-exchanges column chromatography. Isolated ^{211}At-AAMT was smoothly decomposed by oxidation, whereas the *in vitro* stability was enhanced by addition of reducing agent, sodium ascorbate. The LAT1 specificity of ^{211}At-AAMT was confirmed through cell experiments. After tissue distribution of ^{211}At-AAMT was investigated in normal mice, the ^{211}At-AAMT (0.4 Bq/mouse) was injected to nude mice (BALB/c-nu) bearing PANC-1 tumor. The tumor growth was dramatically suppressed by the injection of ^{211}At-AAMT. Thus, we demonstrated the usefulness of the ^{211}At-AAMT for cancer treatment.

3A05 Astatine chemical species in solutions prepared by dry distillation

NISHINAKA, I.[1], HASHIMOTO, K.[2], SUZUKI, H[2]

([1]Tokai Quantum Beam Science Center, QST, [3]Graduate School of Pharmaceutical Sciences, Chiba Univ.)

^{211}At, an astatine radionuclide, with half-life of 7.2 h is one of the prospective candidates for targeted alpha therapy of cancers. Astatine shows some different chemical behaviors in comparison with its homologue iodine. In this work, astatine chemical species have been studied by thin layer chromatography.

The astatine radionuclides ^{208,209,210,211}At and iodine ones ^{120,121,123}I were simultaneously produced by ^{7}Li beam irradiation of a stack of lead and tin targets at the JAEA tandem accelerator facility. The astatine and iodine radionuclides were each separated from the irradiated target with a dry distillation method. No-carrier-added astatine and iodine solutions were prepared using ethanol or water as a solvent. Astatine in the aqueous solution was reacted with an oxidizing or a reducing agent. Separation of the astatine and the iodine ions in the solutions was conducted by thin layer chromatography with an ethanol/water solution. Astatine was separated and identified as anions of At^-, AtO_3^- and AtO_4^-, while iodine was I^-, compared with the standard iodine species.

3A06 Production and distribution of At-211 at RIKEN

SATO, N.[1], HABA, H.[1], YOKOKITA, T.[1], GHOSH, K.[1], WANG, Y.[1], KOMORI, Y.[1], MORI, D. [1], TAKAHASHI, K.[1], KIMURA, T.[2], MATSUMOTO, M.[2]

([1]RIKEN Nishina Center, [2]Japan Radioisotope Association)

Astatine-211 (^{211}At, $T_{1/2}$ = 7.2 h) is one of the promising radionuclides for targeted α-particle therapy. Since 2015, we have produced purified ^{211}At and distributed them to researchers in Japan. In this presentation, we will describe the production technology of ^{211}At and the present status of the ^{211}At distribution at RIKEN.

^{211}At was produced via the ^{209}Bi(α,2n)^{211}At reaction by irradiating a metallic ^{209}Bi target with a 29-MeV α beam from the RIKEN AVF cyclotron. After the irradiation, ^{211}At was purified by a dry distillation method. Radioactivities of ^{211}At were evaluated by γ-ray spectrometry with a Ge detector.

Our thick target yields of ^{211}At are 6.2–7.1 GBq/C at 27.8–28.0 MeV, respectively and they agree with the IAEA recommended values. We confirmed the quantitative ^{211}At production up to the 30-μA beam intensity. The chemical yield of ^{211}At was about 80% with the 0.2-mL methanol as an example. Our ^{211}At products are distributed to about ten research groups in Japan and used in radio-labeling and animal experiments for drug developments.

3A07 Measurement and Estimation of the ^{99}Mo Production Yield by the Acceleration Neutron

TSUKADA, K.[1,2] NAGAI, Y.[2], HASHIMOTO, K.[2], KAWABATA, M.[2,3], MINATO, F.[1], SAEKI, H.[3], MOTOISHI, S.[3], WATANABE, S.[2], ITO, M.[4]

([1]JAEA, [2]QST, [3]Chiyoda technol., [4]Tohoku Uiv..)

We have measured the yield of ^{99}Mo, the mother nuclide of ^{99m}Tc used in nuclear medicine diagnostic procedure, produced by accelerator neutrons. ^{99}Mo was produced by the ^{100}Mo(n,2n)^{99}Mo reaction using neutrons provided by the C(d,n) reaction at the deuteron energy of 40 MeV. The measured yield of ^{99}Mo from a large sample mass of MoO_3 agrees well with the numerical result estimated with the latest neutron data. This result establishes an important finding for the domestic production of ^{99}Mo: approximately 50% of the demand for ^{99}Mo in Japan could be met using a 100 g $^{100}MoO_3$ sample mass with a single accelerator of 40 MeV, 2mA deuteron beams.

3B01 Photoactivity of glass samples characterized by Mössbauer spectroscopy

○ ALI Ahmed [1], NOMURA Kiyoshi [1], AKIYAMA Kazuhiko [1], KUZMANN Ernő [2], HOMONNAY Zoltán [2], KUBUKI Shiro [1]

([1] Graduate School of Science, Tokyo Metropolitan University, [2] Faculty of Science, Eötvös Loránd University)

Slag from municipal household combustion plants acts an environmental problem. Almost all slag is used in road constructing and concrete. Photocatalytic activity has many applications in water splitting and decomposition of organic contaminations. Slag contains iron oxide which is one of the candidate materials for photocatalysis by visible light. The original slag contains 11% of Fe_2O_3. The Fe_2O_3 added slag were prepared by a melt quenching method. The samples were abbreviated as *x*Fe(100-*x*)WS (x=10,30 and 50%). The prepared slag glass samples were further heated at 800°C for 100min. The degradation test of methylene blue dye was used for estimation of photocatalytic activity.

XRD indicate the amorphous structure and crystalline structure before and after heat treatment, respectively. Mossbauer parameters measured at room temperature indicate the presence of $iron^{3+}$ after heat treatment Mossbauer spectra of 50% Fe_2O_3 + Slag showed magnetic components in addition to the paramagnetic components, whereas less than 30% Fe_2O_3 + Slag showed only paramagnetic doublet.

The photoactivity measurements showed the first order rate constant, $K=3x10^{-3}min^{-1}$, while the first order rate constant increased to $17x10^{-3}min^{-1}$ after adding hydrogen peroxide. These results are promising for decomposing organic compound in waste water by using slag as by-product materials.

3B02 Defects formed in ZnO and their contribution to electroconductivity

SHIMIZU, H.[1], SATO, W.[1,2]

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

Zinc Oxide (ZnO) is highly expected for applications to functional devices because of its wide band gap. Commercially available ZnO shows *n*-type conductivity due to different native lattice defects formed during various treatments. It is mentioned that interstitial zinc atoms and oxygen vacancies act as donor defects and zinc vacancies as acceptor defects, and the difference of concentration of each defect corresponds to the net density of charge carriers in the sample; however, little is known about their relationship. In order to investigate the states of lattice defects formed during heat treatment and their contribution to electroconductivity of ZnO samples, in this work, positron annihilation lifetime spectroscopy and conductometry were employed. For the sample annealed at 1,473 K for 1 h in air, positron lifetime was unchanged, whereas electroconductivity was increased compared to the case before the treatment. In spite of the treatment of longer annealing time for 12 h, similar tendencies were observed. From the results for the sample annealed in an oxygen-rich atmosphere, we discuss the mechanism of the occurrence of charge carriers formed in ZnO.

3B03 Nuclear spin relaxation of (^{111}In→)^{111}Cd in Ga-doped Fe_3O_4

SUGIMOTO, T.[1],TAKENAKA, S.[1], FUJISAWA, T.[1], SATO, W.[1,2]

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

In the same way as In ions, impurity Ga ions are also expected to show site-to-site hopping motion in Fe_3O_4, which may lead to charge transportation in the ferromagnetic phase. Thus, we synthesized 0.5 at. % Ga-doped Fe_3O_4, and it was doped with radioactive ^{111}In ions. Their hyperfine fields were investigated in a wide temperature range from 298 to 1073 K by the perturbed angular correlation (PAC) method with the (^{111}In→)^{111}Cd probe. The PAC spectra at 873 to 1073 K ($>T_C$) show relaxation patterns, and it is considered that this nuclear spin relaxation reflects the relative dynamic behavior of the probe nuclei and the extranuclear fields. In the session, this nuclear spin relaxation phenomenon is discussed in relation to the presence of impurity Ga ions.

3B04 Mössbauer Spectroscopic Study on the Chemical Factors Controlling the Electron Hopping Rate in Mixed-valence Trinuclear Iron Complexes

SAKAI, Y.[1], NAKAMOTO, T.[2], OGISO, R.[1], TAKAYAMA, T.[1], KAWASAKI, T.[3], KITAZAWA, T.[3], TAKAHASHI, M.[3]

([1]Daido Univ., [2]Toray Research Center, [3]Toho Univ.)

In mixed-valence trinuclear complexes $Fe^{3+}Fe^{3+}Fe^{2+}O(C_6F_5COO)_6(C_5H_5N)_3 \cdot S$ (hereafter, expressed as “$Fe_3O(C_6F_5COO)_6(C_5H_5N)_3 \cdot S$”), where S is a crystal solvent, iron-valence fluctuation (FeVF) phenomenon was studied mainly by ^{57}Fe Mössbauer spectroscopy, having a time window of ~10^{-7} s, in order to clarify the chemical factors controlling FeVF. In the present work, it was assumed that FeVF would be caused by hopping (or itinerant) of an excess electron among three Fe^{3+} cations in the mixed-valence complexes.

$Fe_3O(C_6F_5COO)_6(C_5H_5N)_3 \cdot S$-complexes were prepared by an exposure method; $Fe_3O(C_6F_5COO)_6(C_5H_5N)_3$ powders were exposed in a vapor of organic compound S including benzene, toluene, cumene, pyridine dichloromethane and so on. Temperature-dependent Mössbauer spectra for different $Fe_3O(C_6F_5COO)_6(C_5H_5N)_3 \cdot S$ complexes were analyzed on the base of electron hopping relaxation to estimate the hopping rates. A Nowick and Wickman’s model on the commercially available software, Moss Winn 4.0 pre, was applied in the present analysis. The relationship between the chemical property of S and the hopping rate was discussed. It was suggested that the high-rate FeVF in the benzene solvate could be facilitated by a molecular rotation of the solvent. The activation energy of electron hopping was estimated to be 3.0 kJ/mol for dichloromethane solvate.

3B05 ^{57}Fe Mössbauer Analysis of Nb doped $SrTiO_3$ Perovskite Oxides

NOMURA, K.[1,2],KUBUKI, S.[1], YAMAKAWA, S.[2], KASARI, M.[2] , IZUNUMA, T[2], KOIKE, Y.[2]
([1] Tokyo Metropolitan University, and [2] Meiji University)

Perovskite oxides are used to control various physical properties such as semiconductor, ferromagnetism, ferroelectricity, superconductivity by element substitution and are therefore candidate materials for next generation devices of spintronics. It is known that the perovskite oxide, $SrTiO_3$(STO), has a band gap of 3.2 eV and a large relative dielectric constant of 300, and that semiconductor characteristics of STO is improved by doping Nb^{5+}. Further doping with magnetic Fe ions is expected to improve the photocatalytic activity in the visible light region and to use as dilute magnetic semiconductor (DMS) at room temperature. STO doped with several % Fe and Nb was prepared by sol-gel synthesis and pyrolysis. The magnetic properties were clarified by a vibration sample magnetometer (VSM). The nanostructure states of the dilute Fe doped STO samples were characterized by ^{57}Fe Mössbauer spectrometry. It is found that the ferromagnetic hysteresis and magnetic relaxation peaks appear with decrease of doped Fe concentration.

3B06 In-Beam Mössbauer Spectra of ^{57}Mn implanted into $LiAlH_4$

SATO, Y.[1], YAMADA, Y.[1], KOBAYASHI, Y.[2,7], KUBO, M.K.[3], MIHARA, M.[4], SATO, W.[5], MIYAZAKI, J[6]., NAGATOMO, T.[7], ANDO, T.[2], TAKAHAMA, N.[2], SOME, K.[2], SATO, M.[2], SATO, S.[8], KITAGAWA, A.[8]
([1]Tokyo Univ. of Science, [2]The Univ. of Electro-Communications, [3]ICU, [4]Osaka Univ., [5]Kanazawa Univ., [6]Hokuriku Univ., [7]RIKEN, [8]NIRS)

In-beam Mössbauer spectra of ^{57}Mn implanted into $LiAlH_4$ were measured at 17, 35, 70, 135, 200, and 300 K to investigate the chemical form of Fe atoms trapped in the $LiAlH_4$ lattice. The experiment was performed using the heavy ion synchrotron accelerator facility (HIMAC) at the National Institute of Radiological Science (NIRS). The Mössbauer spectrum measured at 17 K showed two sets of doublets. The major doublet **A** ($\delta = -0.22$ mm/s, $\Delta E_Q = 0.65$ mm/s) was assigned to FeH_4^-, in which the Fe has an Al substitutional position. The Mössbauer parameters were in agreement with the results estimated by density functional calculations. Another minor doublet **B** ($\delta = -1.5$ mm/s, $\Delta E_Q = 1.1$ mm/s) was assigned to the Fe atom having a Li substitutional position or a position associated with lattice defects. With increasing the temperature of the sample, the intensity of the doublet **B** decreased. This result indicated that the Fe atom was preferentially stabilized at the Al substitutional position having the FeH_4^- form. The intensities of Mössbauer emission changed according to the temperature, and the Debye temperature of the Fe atom trapped in LAH was estimated to be $D = 170$ K.

3B07　Mössbauer Spectroscopic Study on the Chemical States of Iron in Chlorite

HAMASAKA, Y.[1], KAWATA, S.[2], SATO, W.[1,3]
([1]Grad. School Nat. Sci., Tech. Kanazawa Univ., [2]Fac. Sci., Fukuoka Univ., [3]Inst. Sci. Eng., Kanazawa Univ.)

Chlorite has a structure of layers composed of metal ions and silicates, and its chemical formula is generally expressed as $(Mg,Al)_6(Si,Al)_4O_8(OH)_{10}$. Although it is known that Fe^{2+} and Fe^{3+} substitute for Mg and Al in the lattice, there are few reports on their chemical states. In order to elucidate the chemical state of Fe ions in chlorite, in this research, we obtained information on the hyperfine field at the Fe site by ^{57}Fe Mossbauer spectroscopy. Distinct spectra were obtained for chlorite samples taken at different production sites: one has a magnetic component, and the other does not. Assignment of the magnetic component is now underway.

P01 Isothermal gas chromatography of zinc chloride with zinc-65 tracer

MORIYAMA, T.[1], GOTO, S.[1], KUDO, H.[2]

([1]Grad. Sch. of Sci. and Tech., Niigata Univ., [2]Fac. of Sci., Niigata Univ.)

To clarify chemical properties of superheavy elements, gas chromatography of their volatile species has been studied. In general, the chemical properties of superheavy elements are discussed on the basis of the relation between adsorption enthalpies, $\Delta_{\mathrm{ads}}H$, at tracer scale and sublimation enthalpies, $\Delta_{\mathrm{subl}}H$. However, it is difficult to estimate the sublimation enthalpies of superheavy elements accurately because this relation has a large variation. In our laboratory, an adsorption enthalpies of group-4 elements, Zr and Hf, have been measured using isothermal gas chromatography. In this study, an adsorption enthalpy of zinc chloride was measured with the same method for group-4 elements in order to obtain the relation between $\Delta_{\mathrm{ads}}H$ and $\Delta_{\mathrm{subl}}H$ more precisely. Before gas chromatography experiments, evaporation behavior of zinc chloride was researched. First, small quartz wool permeated $^{\mathrm{nat}}$Zn solution of 2 M HCl was placed in a quartz tube. The tube was evacuated, and dried He gas was flushed at about atmospheric pressure. The quartz tube was heated with an electric tube furnace at 100 ˚C for 15 minutes to dry up zinc solution. After that, volatile zinc compound evaporated at higher temperature, and was caught at a water cooling part set at the exit of the tube furnace. Quantifying Zn and Cl, this volatile compound was identified as zinc chloride ($ZnCl_2$). In the presentation, the detailed results with zinc-65 tracer will be discussed.

P02 Anion-exchange behavior of Zr, Hf, and Th in nitric acid for the experiment of Rf

WATANABE, E.[1], NINOMIYA, H.[1], KONDO, N.[1], SHIGEKAWA, Y.[1], YASUDA, Y.[1], OUCHI, K.[1], HAYAMI, S.[2], KASAMATSU, Y.[1], YOKOKITA, T.[3], SHINOHARA, A.[1]

([1]Graduate School of Science, Osaka University, [2]School of Science, Osaka University, [3]RIKEN Nishina Center)

We investigated the time dependence of the anion-excahnge behavior of element 104, Rf, using automated batch-type solid-liquid extraction apparatus (AMBER) to obtain the distribution coefficients (K_d) of Rf in hydrochloric acid. We are interested in the formation of Rf nitrate complex and are planning to obtain the K_d values of Rf by the anion-exchange experiment in HNO_3 using AMBER. In this work, anion-exchange experiments of Zr, Hf (homologues of Rf), and Th (pseudo homologue) in HNO_3 were performed by batch method to search the rapid reaction system and to obtain the comparison data for the chemical study of Rf. Variations of the K_d values of Zr, Hf, and Th were investigated as a function of the shaking time and HNO_3 concentration by using the resins: Muromac ® 1×8 or Aliquat 336. From these results, in the Aliquat 336/HNO_3 system, the anion-exchange reaction equilibrium was achieved by 1-2 minutes of shaking, and it is indicated that this reaction system would be appliable for short-lived ^{261}Rf ($T_{1/2}$ = 68 s) experiment. Based on the present results, we deduced the chemical species of Zr, Hf, and Th, and propose suitable conditions for the anion-exchange experiment of Rf.

P03 Liquid-liquid extraction of Eu and Am with rapid extraction apparatus for experiment of lawrencium

NINOMIYA, H.[1], WATANABE, E.[1], HAYAMI, S.[2], KASAMATSU, Y.[1], SHINOHARA, A.[1]
([1]Grad. Sch. of Sci., Osaka Univ., [2]Fac. of Sci., Osaka Univ.)

In our previous study, we systematically investigated the solvent extraction behaviors of all trivalent lanthanides and actinides except for element 103, laurencium (Lr) in a HDEHP / HNO_3 system. It is difficult to investigate the chemical properties of Lr because of the low production rates and short half-lives of the Lr nuclides. Recently, we developed the flow Injection Solvent Extraction apparatus for superheavy elements (ISE) for online experiments and actually succeeded in studying the liquid-liquid extraction behavior of element 104, Rutherfordium. The purpose of this study is to establish a method to investigate the extraction behavior of Lr. First, we carried out the liquid-liquid extraction experiments of Am and Eu by batch method, and investigated the dependence of the distribution ratios (D) on the concentration of the extractant. In addition, the liquid-liquid extraction experiment of Eu using ISE were conducted under various conditions (the length of the extraction unit : 10, 20, 30 cm, the particle size of the packed chips : 250-500, 500-710, 710-1000 μm). As a result, the D values close to those obtained by batch method (at equilibrium) were acquired by the liquid-liquid extraction experiment using ISE in 0.1-0.5 M HDEHP - Benzene.

P04 Production of ^{236g}Np tracer for determination of ^{237}Np by Accelerator Mass Spectrometry

HAYAKAWA. Y[1], YAMAMORI. K[1], SAKAGUCHI. A[2], SEKIGUCHI. K[2], YANOU. S[3], KOMORI. Y[3], YOKOKITA. T[3], HABA. H[3], SHINOHARA. A[4], YOKOYAMA. A[5]
([1]Graduate School of Natural Science and Technology, Kanazawa University, [2]Mathmatical and science, University of Tsukuba, [3]RIKEN Nishina Center, [4]Graduate School of Science, Osaka Univ., [5]Institute and College of Science and Engineering, Kanazawa University)

A naturally occurring nuclide, ^{237}Np ($t_{1/2} = 2.1 \times 10^6$y), is analyzed with a short-lived Np isotope or Pu considered similar in a chemical behavior, but there is no other tracer that is more satisfactory. Because a long lived isotope of ^{236g}Np ($t_{1/2} = 1.5 \times 10^5$ y) can be used stably and applied to Accelerator Mass Spectrometry (AMS), it has been a candidate as a tracer for years. However, the nuclide has no established production method. In this study, nuclear reactions of U + p and Th + Li ware carried out to produce Np isotopes and measure their yields in γ-ray spectrometry with a Ge detector. It was found that the ^{234}Np, ^{235}Np and ^{236m}Np nuclides are produced in the reactions. Compared to the U + p reaction, cross sections of Np isotopes in the Th + Li reaction are smaller, but it can be expected that the formation ratio of ^{236g}Np to ^{237}Np interfering with its measurement in nature is better. Based on the results, we are studying the optimum condition of tracer production of ^{236g}Np.

P05 Development of a Monte Carlo simulation code for analyses of adsorption and desorption behavior of short-lived isotopes in a surface ion-source

TOKOI, K.[1,2], SATO, T.K.[1], EICHLER, R.[3], ITO, Y.[4], TOMITSUKA, T.[1.3], SUZUKI, H.[5], ASAI, M.[1], TSUKADA, K.[1], TOYOSHIMA, A.[1], CHIERA, N.M.[1], NAGAME, Y.[1,2]

([1]JAEA ASRC, [2]Graduate School of Science and Engineering, Ibaraki Univ., [3]PSI, [4]Graduate School of Science and Technology, Niigata Univ., [5] College of Science, Ibaraki Univ)

Superheavy elements (SHEs) can be synthesized in heavy-ion nuclear fusion reactions using accelerators. However, due to the typically low production rates and short half-lives of SHEs, few information on their electronic structure and on their chemical properties is available.

We developed a new experimental method combining vacuum chromatography and surface ionization, and applied this to investigate adsorption behavior of lawrensium (Lr,Z =103) on a surface of high temperature metallic tantalum. The adsorption behavior of Lr was similar to that of lutetium (Lu, Z = 71), showing a low volatility in relation to other lanthanides.

In this work, a Monte Carlo simulation code has been newly developed to study ionization and chromatographic behavior of short-lived atoms. In the presentation, we will report the details of a developed simulation code and calculation results.

P06 Hydroxide precipitation experiment of Mg, Ca, Sr, Ba, and Ra for chemical study of Nobelium

NAGASE, M.,[1] KASAMATSU, Y.,[2] KONDO, N.,[2] NINOMIYA, H.,[2] WATANABE, E.,[2] SHINOHARA, A.[2]

Nobelium (No) belongs to actinide series, and the most stable ion valence of No in aqueous solutions is reported to be +2 although those of the other heavy actinide elements (and all lanthanides) are all +3. However, the chemical experiments of No were few because of low production rates and short half-lives of the No nuclides. It was reported that the chemical behaviors of alkaline earth metals are similar to that of No in some extraction experiments. The purpose of this study is to establish the experimental mehod to investigate the precipitation properties of No by applying the coprecipitation method with samarium (Sm) hydroxide. In this work, we investigated the hydroxide precipitation behaviors of alkaline earth metals (macro amounts) by conventional filtration method. Then, we applyed the coprecipitation method with Sm hydroxide to ^{85}Sr, ^{133}Ba, and ^{226}Ra carrier-free tracers. The precipitation yields were obtained from each metal concentration in precipitate, filtrate, cup, and standard samples. The precipitation yields increased with higher hydroxide ion concentration, and the yield of heavier element increased at higher hydroxide ion concentration. This tendency is consistent with the order of solubility product of these elements: typical properties are observed in the precipitation experiment. However, the coprecipitation yields with Sm did not show the same tendency.

Polynuclear species might be formed in the precipitation experiment using macro amounts of metals.

P07 Development of Si-Ge detector array for identification of superheavy nuclide using X-ray measurement

KAJI, D. [1], MITSUOKA, S. [1,2], HIRANO, T. [1,2], NIWASE, T. [1,2], MORIMOTO, K. [1]

([1]RIKEN, Nishina Center for Accelerator-Based Science, [2]Kyushu University, Graduate School of Science)

Superheavy (SHE) element device development team in RIKEN has developed a new focal plane detector to study on production and decay properties of SHE nuclide using a gas-filled recoil separator GARIS-II. Conventional identification method of SHE nuclide was based on the genetic relation between mother and daughter, was obtained from measuring the α-decay and spontaneous fission (SF) by Si-box detector. So, detailed decay scheme including γ-decay could not be obtained. The new detector array is also useful as a probe for detection of X-ray coincided with α-decay. Observation of X-ray originating from SHE nuclide is one of important criteria that must be satisfied for the discovery of a new chemical element to be recognized. We evaluated basic properties of the Si-Ge detector array using nuclear reactions of ^{51}V + ^{nat}La. In this report, the results on the first α-γ(X) spectroscopic study at the focal plane of GARIS-II will be given in detail. (152 words)

P08 Extraction behavior of Rf from HCl to Aliquat 336 in $CHCl_3$ or CCl_4

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.,1 HABA, H.,3 KUBOKI, Y.,3 KOMORI, Y.,3 YOKOKITA, T.,3 YANO, S.,3 SATO, N.,3 SHINOHARA, A1.

The purpose of this study is to investigate the liquid-liquid extraction behavior of Rf to Aliquat 336 from HCl. In our previous study, we carried out the liquid-liquid extraction experiment of Zr and Hf as a model experiment of Rf, and developed the flow Injection Solvent Extraction apparatus for superheavy elements (ISE). Then, the extraction behavior of Rf from HCl to Aliquat 336 $CHCl_3$ solution was also investigated using ISE. In this work, we conducted the extraction experiment of Rf to Aliquat 336 CCl_4 solution. ^{261}Rf ($T_{1/2}$ = 68 s) and ^{169}Hf ($T_{1/2}$ = 3.24 min) were produced in the ^{248}Cm(^{18}O,5n)^{261}Rf and ^{nat}Gd(^{18}O,xn)^{169}Hf reactions, respectively, at the RIKEN AVF cyclotron. The aqueous phase was 9.3 M HCl. In the experiment, the nuclear reaction products were transported by the He/KCl gas-jet system to a chemical laboratory, and were dissolved in aqueous solution with a dissolution apparatus. The aqueous and organic solutions were injected to the ISE. After that, we collected two phases separately on different Ta plates and subjected to α-particle measurement. As a result, we obtained 55 α counts of ^{261}Rf and ^{257}No and acquired the *D* values of Rf. Although the *D* valuse of Rf using $CHCl_3$ showed clearly defferent behavior from those of Zr and Hf, the *D* values obtained in this work were similar to those of Zr, suggesting similar results to that in the previous anion-exchange experiment of Rf.

P09 Cation exchange behavior of lanthanide and actinide using α-HIBA

KASHIHARA, A.[1], SUEKI, K.[1], TSUKADA, K.[2], TOYOSHIMA, A.[2], NINOMIYA, H.[3]
([1]Graduate School of Science, Tsukuba Univ., [2]JAEA, [3]Graduate School of Science, Osaka Univ.)

Recently, JAEA obtained einsteinium (Es)-254 from Oak Ridge National Laboratory. The Es-254 material is required to be separated from its daughter nuclides and impurities to use as a target for its physical and chemical experiments. Although Es is isolated from lanthanide and other actinides by means of cation-exchange chromatography in α-hydroxy-isobutyric acid (α-HIBA) solution, its ionic radius, affecting effective charge and thus stability constants, has not been determined yet. Thus, in the present study, we carried out cation-exchange chromatography of heavy actinides in α-HIBA to deduce ionic radius of Es from the relationship between ionic radius and the elution position. We first examined peak positions of lanthanide elements in the elution chromatography to ensure the relationship for lanthanide. Peak positions of Fm, Es, Cf, and Am were then measured in the chromatography with α-HIBA, and a similar trend as lanthanides was found in the relationship for actinides. We are now obtaining further elution data of Cm and Bk to establish the relationship for actinide.

P10 Thermal-chromatography of astatine compounds in gas-phase generated in dry-separation process

ICHIMURA, S. [1], TOYOSHIMA, A.[2], ZHANG, Z. [1], IKEDA, T. [1], OOE, K.[3], NAGATA, K.[2,4], YOSHIMURA, T.[2,4], SHINOHARA, A. [1,2]
([1]Graduate School of science, Osaka Univ., [2]Institute for Radiation sciences, Osaka Univ., [3]Graduate School of medicine, Osaka Univ., [4]Radioisotope Research Center, Osaka Univ.)

Recently, targeted radiation therapy using an α-particle-emitting nuclide, ^{211}At, is attracting a great deal of attention. In our research group, experiments such as drug synthesis and animal tests are conducted using ^{211}At solution prepared by dry distillation. In this method, an irradiated Bi target containing ^{211}At is first heated in a closed system and then ^{211}At is vaporized and carried by a carrier gas to a cold trap. ^{211}At is accumulated in the cold trap and then dissolved in a small amount of solution which is then supplied to the following experiments. However, the recovery yield of ^{211}At is strongly dependent on the composition of the carrier gas. This is probably because chemical species of ^{211}At and their ratios are different for the gas conditions. In this work, we newly developed a thermal-chromatography system and investigated the adsorption behavior of gaseous ^{211}At compounds on quartz surface to study generated chemical species of ^{211}At. As a result, the adsorption behavior of ^{211}At is different for each gas-composition. This suggests that several types of ^{211}At compounds with different volatilities are produced at the same time. Details of the adsorption behavior will be presented in the meeting.

P11 Coincidence measurement of the beta-particle and the electron from Ac-229 for observing the internal conversion electron of Th-229m

SHIGEKAWA, Y.[1], KASAMATSU, Y.[1], YASUDA, Y.[1], KONDO, N.[1], NINOMIYA, H.[1], WATANABE, E.[1], HAYAMI, S.[1], SHINOHARA, A.[1]

([1]Graduate School of Science, Osaka University)

The excitation energy of ^{229m}Th is as low as 7.8 ± 0.5 eV. It has been expected that the decay process and the half-life of ^{229m}Th varies depending of the chemical environments. Although the internal conversion (IC) electron of ^{229m}Th was recently detected and the IC half-life was found to be around 7 μs, the precise excitation energy is yet to be measured. We are aiming to determine the precise excitation energy by measuring the energy of the IC electron of ^{229m}Th. In this work, we tried to measure the IC electron of ^{229m}Th with the coincidence measurement of the beta particle and the electron from ^{229}Ac. With the retarding field between the ^{229}Ac sample and the electron detector, we can measure the energy of the IC electron. First, we fabricated ^{229}Ac by irradiating 20-MeV proton to ^{232}Th. Then we purified ^{229}Ac and prepared an electrodeposited ^{229}Ac sample. We measured the detection time of the beta particle and the electron from the sample, and obtained coincidence spectra. The purification of ^{229}Ac from ^{232}Th and fission products well succeeded. However, the signal which corresponds to the disintegration of ^{229m}Th was not detected. We need to find the method for preparing the extremely thin sample which enables the IC electron to escape from the surface.

P12 Verification of excitation functions for photonuclear reactions with simple irradiation system

KIKUNAGA, H.[1], KASHIWAGI, S.[1], TAKAHASHI, K.[1], NANBU, K.[1], TSUKADA, K.[1], NODA, S.[2], IDA, H.[2], IMURA, R.[2], OURA, Y.[3]

([1]ELPH, Tohoku Univ., [2] JFE Engineering Corporation, [3]Graduate School of Science, Tokyo Metropolitan Univ.)

Radioisotope production by using bremsstrahlung irradiation has been carried out at ELPH, Tohoku Univ. The production yields of photonuclear reactions are estimated by multiplying bremsstrahlung spectrum and excitation functions. The former can be reproduced well by particle transport simulation codes such as PHITS code, while the latter is difficult to obtain both experimentally and theoretically. Therefore, even (g,n) reactions, there are not many measurements of the excitation functions and its values differ from experiment to experiment. In this study, we try to verify the literature values of the excitation functions and those obtained by a calculation code by using a simple activation method.

P13 Network calculation of p-nuclei in IaSN using Hauser-Feshbach statistical calculation

KINOSHITA, N

(Shimizu Co.)

The type Ia supernovae (IaSN) take place when a mass exceeds the Chandraskhar limit. Nuclear reactions with photon, proton, and neutron in the explosive environment produce numerous nuclides including neutron-deficient nuclei, p-nuclei. Cross sections of these nuclear reactions depend on a theoretical calculation using the Hauser-Feshbach (HF) statistical model. Some parameters such as optical model potential and level density affect strongly the precision of the cross sections. The parameters were optimized by comparing experimental cross sections reported by several researchers with the theoretical cross sections for which different parameters were applied. Then, cross sections of 3052 nuclei from C up to Cm with photon, proton, and neutron were calculated by applying both default parameter and the optimized parameters in the HF calculation. The network calculation was done using reaction rate calculated by these 2 different parameters in the HF calculation. Cross sections of alpha emission reaction strongly depend on the optical potential of alpha particle. Therefore, the cross sections estimated with default input parameter are larger than those estimated with the modified input parameter in the theoretical calculation. The different cross sections result in different pathway for the production of the p-nuclei. I discuss the production of the p-nuclei affected by the different input parameter.

P14 Concentrations of ^{129}I in river waters of eastern Fukushima, Japan

UEDA, S.[1], OCHIAI, S.[2], HASEGAWA, H.[1], KAKIUCHI, H.[1], SATOH, Y.[1], OHTSUKA, Y.[1]
HISAMATSU, H.[1]

([1]Department of Radioecology, Institute for Environmental Sciences, [2]LLRL, Institute of Nature and Environmental Technology, Kanazawa Univ.)

River waters in the Ukedo, Odaka, Ohta, Niida, Mano Rivers etc. located in the eastern part of Fukushima Prefecture were collected in August 2016 and August 2017, and concentrations of ^{129}I in these river waters were measured by AMS. The maximum value of ^{129}I concentration in river water was 1.6E-6 Bq L^{-1} of the Ukedo River, and the minimum value was 2.4E-7 Bq L^{-1} of the Hirose River. The range of ^{129}I/^{127}I atomic ratio in river water was 3.9E-9 to 7.4E-8, and the maximum value was the Ukedo River both 2016 and 2017. There was also a significant correlation between ^{129}I/^{127}I atomic ratio and dissolved ^{137}Cs concentration in river water. Therefore, it is presumed that the amount of ^{129}I and ^{137}Cs deposited in the catchment area of the river in the eastern part of Fukushima Prefecture has a relatively similar distribution.

P15 Examination of Chemical States Analysis Condition of Heavy Metals in Municipal Solid Waste Incineration Fly Ash Containing Radioactive Cesium

TAKAHASHI, T.[1], MIZUNUMA, T.[2], KASARI, M.[2], KOIKE, Y.[1]
([1]Sch. of Sci. and Tech., Meiji Univ., [2]Grad. Sch. of Sci. and Tech., Meiji Univ.)

MSWI fly ash was sampled in Fukushima Prefecture.

Radioactive cesium (^{134}Cs and ^{137}Cs) and toxicity heavy metals were often contained in a municipal solid waste incineration (MSWI) fly ash. Radioactive cesium was concentrated in the MSWI fly ash incinerated by high temperature. Several studies reported on radioactive nuclides including radioactive cesium in MSWI fly ash were focused on its speciation. It is necessary to investigate the chemical states for the purpose of evaluating the toxicity of not only radioactive cesium but also other heavy metals. In this study, chemical states analysis conditions were optimized to understand elution behavior of heavy metals in MSWI fly ash. Thus, the sequential extraction test based on method of Tessier *et al.* was conducted using simulated fly ash samples mixed with silicon dioxide and some lead compounds. For example, according to the leaching test with pure water for a model sample of fly ash containing lead (II) chloride, a part of lead was not eluted. An optimal sequential extraction procedure for MSWI fly ash will be proposed by evaluating the analysis result of the model samples of fly ash.

P16 On-site determination of radioactive cesium in environmental sample using portable NaI scintillation counter.

MOCHIMARU, T.[1], AKIYAMA, M.[2], HAGIWARA, K.[1], KOIKE, Y.[1]
([1] Sch. of Sci. and Tech., Meiji Univ., [2] Grad. Sch. of Sci. and Tech., Meiji Univ.)

An on-site determination method for radioactive cesium (^{134}Cs and ^{137}Cs) in environmental sample has been developed using a portable NaI scintillation counter combined with solid phase extraction disk. By using potable measuring equipment, rapid on-site analysis is possible without transporting the collected environmental sample to the laboratory. Radioactivity of radioactive cesium in the disk-shaped radioactive standard materials (IAEA) and solid phase extraction disk concentrated soil eluate were determined by using NaI scintillation counter, Get Smart XR, and HP-Ge spectrometer. Radioactive cesium was identified in each of the gamma-ray spectra obtained by the two spectrometers. Detection efficiencies for Get Smart XR were calculated using area of photo peak in gamma-ray spectra and concentration of ^{137}Cs in the soil eluate. Relative errors between activity concentration of ^{137}Cs by Get Smart XR and HP-Ge spectrometer were suppressed to ± 5%. The present method using the portable NaI scintillation counter will be applicable to rapid on-site determination for radioactive cesium in environmental samples.

P17 ICP-MS analysis of river water and treated sewage targeted on gadolinium and iodine fractions due to contrast mediums of MRI imaging and X-ray CT inspection, and correlation of their concentrations and other mental elements and naturally occurring radioactive nuclides

MATSUMOTO-KAWAGUCHI, E[1]., IMAGAWA, A[1]., MIYAMOTO, N[1]., SAKAMA, M[1]., FUKUSHI, M[2]., INOUE, K[2]., SATO, T[1]., IHARA, T[1]., IMOTO, S[1].

([1]Graduate School of Health Sciences, Tokushima Univ., [2]Graduate School of Human Health Sciences, Tokyo Metropolitan Univ.)

A purpose of this study is to investigate the relevance with level characteristics of gadolinium and iodine concentrations to contain in river waters with the MRI and iodine contrast mediums such as X-ray computed tomography and the angiography by analyzing the river water samples which we had collected regionally. We have measured 26 element concentrations among the river water samples, which supplied to us at the part of the collaboration program together with Fukushi and others of Tokyo Metropolitan University and also which they had collected from 30 sampling points of the Tonegawa basin using our ICP-DRC-MS apparatus (Tokushima Univ.), and investigated those distribution behaviors ranged from the upstream to downstream. Furthermore, we have examined the correlation between the location of the sewage disposal plants and the concentration properties of gadolinium and iodine in those river samples. These results confirmed that the characteristics of the concentration level behavior of gadolinium from upstream to downstream on the river basin represented similar tendency like the element group of the artificial pollution such as aluminum and zinc.

P18 The Geochemical cycles at the surface in the Sea of Japan implied from lateral profiles and seasonal variations of ^{228}Th/^{228}Ra and ^{234}Th/^{238}U activity ratio

MOROKADO, T[1]., INOUE, M[1]., SHIROTANI, Y[1]., FUJITA, A[1]., HANAKI, S[1]., MORITA, T[2]., MIKI, S[2]., HONDA, N[3]., HIROHASHI, N[4]., ANDO, H[5]., SATO, Y[6]., MORIWAKI, K[6]., NAGAO, S[1].

([1]LLRL, Kanazawa Univ., [2]NRIFS, FRA., [3]JSNFRI, FRA., [4]Shimane Univ., [5]Niigata Univ., [6]Shimane Prefectural Government.,)

The radium isotopes (^{226}Ra, $t_{1/2} = 1600$ y; ^{228}Ra, $t_{1/2} = 5.75$ y) are dissolved in, and circulates with seawater in marine environment. Those main source of supply are coast sediment and continental shelf. Then ^{228}Ra/^{226}Ra ratio in coastal area is higher than open sea influenced by short life of ^{228}Ra. On the other hand, the thorium isotopes (^{228}Th; $t_{1/2} = 1.91$ y, ^{234}Th; $t_{1/2} = 24$ d) are adsorb suspended particle and scavenged in marine environment. Therefore, compare with each parent nuclides, ^{228}Th/^{228}Ra and ^{234}Th/^{238}U ratios can be the indexes of particle and adsorption materials. In this study, we discus on the circulation and scavenging effect of Sea of Japan by horizontal distribution and seasonal variation of ^{228}Ra/^{226}Ra、^{228}Th/^{228}Ra and ^{234}Th/^{238}U ratio implying high-resolution spatial researches.

The sea water samples were collected from 2014 to 2018 at the Sea of Japan Honshu coast. Each samples are 20L non-filtered surface water.

P19 Distribution of I-129 in forest soil from the boundary of Fukushima Daiichi Nuclear Power Plant

YANG, G.[1], TAZOE, H.[1], TSUKADA, H.[2], YAMADA, M.[1]
([1]Institute of Radiation Emergency Medicine, Hirosaki Univ., [2]Institute of Environmental Radioactivity, Fukushima Univ.)

Long-lived ^{129}I ($t_{1/2}$ = 1.57×10^7 y) can be used to reconstruct the early dispersion and distribution of the Fukushima Daiichi Nuclear Power Plant (FDNPP) derived ^{131}I ($t_{1/2}$ = 8.03 d) in the environment, and to study the behavior of radioiodine after deposition onto terrestrial system. The core soil samples were collected from forest at about 1.1 km south from the FDNPP in 2017, and the distribution of ^{129}I from the FDNPP accident was determined by the combination of TMAH extraction and ICP-MS/MS analysis. The surface soil samples (0–5 cm depth) were heavily contaminated with ^{134}Cs (262–1,588 Bq g^{-1}) and ^{137}Cs (259–1,570 Bq g^{-1}) (decay corrected to March 11, 2011). The ^{129}I activities decreased sharply with the soil depth, from 1,211 to 34.1, 534 to 78.9, and 2,536 to 51.3 mBq kg^{-1}, respectively, for the three sites. In addition, the ^{129}I inventories were observed to be 35.0±1.0 (0–15 cm depth), 28.2±0.9 (0–17.5 cm depth), and 55.1±1.8 (0–15 cm depth) Bq m^{-2}, respectively.

P20 Low-levels of the FDNPP-derived radiocesium of marine products from coastal areas in the Sea of Japan

INOUE, M.[1], YAMASHITA, S.[1], TAKEHARA, R.[1], MIKI, S.[2], NAGAO, S.[1]
([1]LLRL, Kanazawa Univ., [2]NRIFS-FRA)

Low-background γ-spectrometry combined with the ^{40}K-removal treatment technique was applied to marine products collected in coastal areas of the Sea of Japan during 2012–2017. The radiocesium concentrations in the samples were < 0.005–0.02 Bq/kg-wet for ^{134}Cs and < 0.05–0.17 Bq/kg-wet for ^{137}Cs. The concentration factors (CFs) of ^{134}Cs and ^{137}Cs in fishery products were estimated to be ~30–100, which approximately agreed with those of ^{137}Cs recorded before the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident around the Japanese Archipelago. The uptake of ^{134}Cs from local sources (e.g., ^{134}Cs-contaminated coastal sediments and the leakage from nuclear power plants) along Honshu was not evident in the marine products. Therefore, it was concluded that marine products have continually taken up ^{134}Cs from the ambient seawater and prey organisms (through the food chain) in the inhabitant areas after the re-entry of ^{134}Cs with the Kuroshio Current water from the Pacific Ocean-side (after 2013).

P21 Root Uptake Effect on Cs-137 Concentration in Aquatic Plants

TAGAMI, K.[1], UCHIDA, S.[1]

([1]QST-NIRS)

When a nuclear accident occurred, radiocaesium is one of the most important radionuclides among the discharged radioactive materials to the environment because it can cause a major radiation source to humans several years after the accident. In the terrestrial environment, soil and water can be radiocesium sources to plants. Two to three years after the Fukushima Daiichi nuclear accident, water soluble radiocaesium concentrations in fresh waters became low, therefore, in the upland conditions, soil is the main source to contribute radiocaesium concentrations in crops. In freshwater conditions, e.g, rivers and lakes, soluble radiocaesium still may affect the concentrations in emergent plants because they can take up radiocaesium through their aboveground part. However, it was not clear which is the major source to the plants, soil or water. In this study, therefore, uptake of Cs-137 by emergent plants (literature data) was examined by comparing their Cs-137 uptake with those of submerged and floating plants (literature data) as well as upland herbaceous plants (field observation data). From the results, it was estimated that around 20% of the total Cs-137 in emergent plants was from water.

P22 Air dose rate monitoring at the portable radiation monitoring stations in Fukushima Prefecture

MIYATA, S., TANAKA, H., SATO, S., OTA, Y., ISOGAI, K.

(Japan Chemical Analysis Center)

Air dose rate monitoring have been carried out with portable monitoring stations (Fuji Electric Co., Ltd.) at 80 locations in Fukushima Prefecture since April 2012. 60 monitoring stations have 2 detectors, silicon semiconductor detector for high air dose rate and NaI (Tl) detector for low air dose rate, and 20 monitoring stations have only silicon semiconductor detector. Decreases of air dose rates were observed at all the monitoring stations due to the decay of artificial radioisotopes (^{134}Cs and ^{137}Cs). In addition to the long-term dose reduction trends, short-term fluctuations in air dose rates were observed, which were caused by various natural and artificial factors. Artificial factors include changes in surrounding environment such as “decontamination activity around the monitoring station”. Regarding artificial factors, presence of radioactive wastes around the station will also cause the increase of air dose. At some monitoring stations, abrupt changes in dose rates were observed due to this type of factors. Regarding natural factors, it is generally known that the rainfall causes the increase of air dose due to the washout and rainout of Radon progenies. However, decrease of air dose rate while rainfall were surrounded by the area with relatively high radioactivity concentration. This is caused by the water layer formed on the ground surface which functions as a shield against the gamma-rays from ^{134}Cs and ^{137}Cs in the ground, similar to the shielding effect by snow cover.

P23 Determination of platinum group elements of GPt-5 chromitite reference sample by using INAA and ID-ICP-MS

SHIRAI, N.[1], AKHTER, R.[1a], EBIHARA, M.[1b]

([1]Graduate School of Chemistry, Tokyo Metropolitan University, Present affiliation: [a]Bangladesh Atomic Energy Commision, [b]Waseda University)

Major, minor and trace element (Mg, Al, Ca, Sc, V, Cr, Mn, Fe, Co, Ni, Zn and Ir) abundances for a chromitite reference sample, GPt-5, were obtained by using INAA. Except for Ni, our INAA data are in good agreement with literature values. By using ID-ICP-MS coupled with NiS fire-assay pre-concentration, the same samples previously analyzed by INAA were used for the determination of platinum group elements (Ru, Rh, Pd, Os, Ir and Pt) in order to avoid the sampling bias in assessing the effectiveness of the NiS fire-assay pre-concentration. It was possible to determine Ir abundance by using both the INAA and ID-ICP-MS, and the values obtained were consistent with each other within the measurement precision. It was confirmed that the NiS fire-assay pre-concentration applied in this study can digest chromitite and that the accurate determination of platinum group element for chromite-containing rocks can be performed by using our analytical method. Based on the Ru/Rh ratio, GPt-5 chromitite reference sample is classified as a podiform chromitite.

P24 Depth profile of 137Cs concentration in soil collected in 2016 near the Fukushima Dai-ichi Nuclear Power Plant

NINOMIYA, K.[1], KITA, K.[2], SHINOHARA, A.[1], KAWATSU, K.[3], MINOWA, H.[4], FUJITA, N.[1], OHTSUKI, T.[5], TAKAMIYA, K.[5], KINO, Y.[6], KOARAI, K.[6], SAITO, T.[7], SATOU, Y.[8], SUEKI, K.[9], TAKEUCHI, Y.[10], DOI, T.[10], CHIMURA, K.[10], ABE, Y.[11], INAI, Y.[1], IWAMOTO, Y[12], UESUGI, M.[13], ENDO, S.[14], OKOCHI, H.[12], OKUMURA, S.[15], ONO, T.[11], ONOSAKI, H.[11], KATSUMI, S.[12], KANDA, A.[1], KINNO, S.[12], THANH, N. T.[14], KUBO, K.[16], KOIKE, Y.[15], MOTOOKA, A.[12], SUZUKI, M.[6], SUZUKI, K.[3], SUZUKI, A.[1], TAKASE, T.[3], TAKAHASHI, M.[1], TAKENAKA, S.[13], ZHANG, Z.[1], NAKAI, I.[11], NAGAO, S.[13], NAKAMURA, S.[14], NANBU, A.[1], NISHIYAMA, J.[6], NISHIYAMA, Y.[5], FUKUDA, D.[15], FUJII, K.[15], MIYAZAWA, N.[13], MURANOI, T.[6], MORIGUCHI, Y.[17], YATAGAI, A.[18], YAMAMORI, K.[13], YOKOYAMA, A.[13], YOSHIDA, G.[19], YOSHIMURA, T.[1], WATANABE, A.[3]

([1]Osaka Univ., [2]Ibaraki Univ., [3]Fukushima Univ., [4]The Jikei Univ. School of Medicine, [5]Kyoto Univ., [6]Tohoku Univ., [7]Shokei Gakuin Univ., [8]JAEA, [9]Tsukuba Univ., [10]National Institute for Environmental Studies, [11]Tokyo Univ. of Science, [12]Waseda Univ., [13]Kanazawa Univ., [14]Hiroshima Univ., [15]Meiji Univ., [16]International Christian Univ., [17]The Univ. of Tokyo, [18]Hirosaki Univ., [19]KEK)

Soil sampling up to 30 cm depth at 31 points near the Fukushima Dai-ichi nuclear power plant was conducted in 2016. After drying the sample, ^{134}Cs and ^{137}Cs concentration with each 2.5 cm depth was determined from gamma-ray spectrometry. The depth profiles of the ^{134}Cs and ^{137}Cs concentration were investigated. Most sites have the highest concentration in the surface samples. The result shows ^{137}Cs still remains near the soil surface region though over 5 years was passed from the Fukushima Dai-ich Nuclear Power Plant accident.

P25 Decontamination of paddy field soil using a stable isotope

DEZAWA, Y.[1], MIYOSHI, H.[2], HIGAKI, S.[3] , MORI, K.[4], SERA, K.[5], YANAGA, M.[6]

([1]Grad. School of Inte. Sci. and Tech., Shizuoka Univ., [2]ARREM, Tokushima Univ., [3]ISC, Tokyo Univ., [4]ING Co., Ltd, [5]Cyclo. Res. Center, Iwate Medical Univ., [6] Fac. of Sci., Shizuoka Univ.)

A large amount of radioactive material was released into the environment by the Fukushima nuclear power plant accident. Paddy field soil was also contaminated with radioactive cesium. Although there have been no reports that activity of rice produced in Fukushima prefecture exceeding the reference value was detected recently, decontamination is necessary to prevent damage by rumors. We previously reported that the absorption of radioactive cesium from artificially contaminated soil into rice plants increased by adding a stable isotope to irrigation water. In this study, cultivation tests were conducted by using paddy field soil collected in Fukushima prefecture to examine whether the absorption of radioactive cesium from really contaminated soil increase or not. In addition, the test was conducted by changing the amount of the stable isotope to be added. Harvested brown rice was transferred to a U8 container and radioactivities of ^{137}Cs and ^{134}Cs in the brown rice were measured with a high purity Ge semiconductor detector. As a result, in groups which added Cs stable isotope, the absorption amount increased compared with the group which was not added.

P26 Measurements of Cs-137 activity concentration in individual organs of wild raccoons in Namie town

TAKUMI, O.[1], KAZUMA, K.[1], YASUSHI, K.[1], HIROAKI, T.[1], TOSHITAKA, O.[1,2], ATUSHI, T.[3], TOSHIHIKO, S.[4,5], YOSHINAKA, S.[4], MIREI, C.[4], YOHEI, F.[6], VARERIE GOH SEE TING.[6], KENTARO, A.[7], AKIFUMI, N.[8], MASATOSHI, S.[9], HIDEAKI, Y.[10], TOMISATO, M.[6,7], TSUTOMU, S.[1,2], HISASHI, S.[4]

([1]Depertment of Chemistry, Tohoku Univ., [2]Institute for Excellence in higher Education, Tohoku Univ., [3]Tohoku University hospital, Tohoku Univ, [4]Graduate school of Dentistry, Tohoku Univ, [5]International Research Institute of Disaster Science, Tohoku Univ., [6]School of Health Science, Hirosaki Univ., [7]Institute of Radiation Emergency Medicine, Hirosaki Univ., [8]Faculty of Pharmaceutical science, Hokkaido University of Science. [9]Institute for Disaster Reconstruction and Regeneration Research, Tohoku Univ., [10]Faculity of Agriculture, Niigata Univ.)

We measured Cs-137 activity in individual organs of wild raccoons in Namie town to investigate Cs-137 distribution in raccoon. We caught twelve raccoons with a box trap, and collected femoral muscle, liver, kidney, spleen, heart, lung, and blood together with the samples for biological measurement. We found a weak positive relationship between air dose rate area of the sampling site and Cs-137 activity concentration in femoral muscle in high air dose rate area, and did not found the correlation in low air dose rate area. Cs-137 activity concentrations in femoral muscle did not always reflect the air dose rate of the sampling site because raccoons can live in both city and mountain area. This result indicates an importance of the measurements of individual raccoons. Activity concentration of femoral muscle was the highest of all the organs measured, and activity concentration of blood was lowest. Activity concentrations of the other organs (liver, kidney, spleen, heart, lung) ware almost half value of the femoral muscle.

P27 Recent in Environmental Tritium Concentrations and Actual Analytical Measurements

SHIMA, N.[1], TAMARI, T.[1]

([1]Kyushu Environmental Evaluation Association)

We investigated the tritium concentration in the current environment in Kyushu district. Each precipitation and monthly atmospheric water vapor were collected at Fukuoka City from Feb 2016. Seawater and river water were collected at Fukuoka, Saga, Oita, Kumamoto, Miyazaki, Kagoshima in 2017. Every samples were performed electrolytic enrichment and measured by low background LSC. Tritium concentrations in precipitation and atmospheric water vapor were in the range of 0.08 to 1.25 Bq / L and 0.19 to 0.88 Bq / L, respectively. The tritium concentrations in the seawater ranged from 0.05 to 0.15 Bq / L. Tritium concentrations in the river water ranged from 0.16 to 0.27 Bq / L. In recent years, tritium concentrations in environmental water in the Kyushu district are very low, and it is suggested that electrolytic enrichment for sample water is indispensable to evaluate its concentration.

P28 Determination of ^{134}Cs by Sum peak method using Ge semiconductor detector

SUZUKI, K., AKIYAMA, M., OTA, Y., ISOGAI, K.

(Japan Chemical Analysis Center)

An attempt was made to expand the application of the 'sum-peak' method to determine ^{134}Cs in environmental samples. Coaxial type Ge detectors and cylindrical sample containers were used for gamma-ray measurement.

Firstly, ^{134}Cs point source was measured in the vicinity of the Ge detector and the activities were calculated by 'sum-peak' method. The calculated activities were in good agreement with the spiked value within 10% in the measurement geometry that the distance between the source and the detector endcap is less than 5cm. Secondly, ^{134}Cs volume sources with different packing thickness to the containers were measured on the endcap of the detectors. From the results, geometry dependence of the calculated values by 'sum-peak' method were evaluated. Finally, airborne dust samples collected in Chiba city several months after the Fukushima Daiichi Nuclear Plant Accident on 2011 were measured and ^{134}Cs activities were calculated by 'sum-peak' method. The results agreed within 10% with the activity values determined by conventional calibration method. This shows that the 'sum-peak' method is applicable at least under limited measurement conditions.

P29 Estimation of aerosol attaching to unnamed aerial vehicle body surface flying in radioactive plume

OSADA, N.[1], ISHIZAKI, A.[2], NISHIZAWA, Y.[3], YUUKI, Y.[3], SANADA, Y.[2]
([1]Advanced Science Research Center, Okayama Univ., [2]Japan Atomic Energy Agency, [3]OYO corp.)

Unmanned aerial vehicle to survey the radioactivity of the ground surface is one of useful procedure to environmental radiation. When the aerial vehicle will fly in the radioactive plume, the radioactive aerosol attach to the aerial vehicle body surface. The attaching radio-materials will disturb the survey. In this study, we estimated attaching amount of the radioactive aerosol to the aerial vehicle body surface for evaluation of the disturbance. Two model aerial vehicles were set in constant wind including potassium solution aerosol. The attached radio-materials were collected by a filter paper, and measured with a Ge-semiconductor detector. The recovered radio-materials were also analyzed by atomic absorption spectroscopy. The result suggests that the velocity of attachment was from 2.0×10^{-4} to 9.7×10^{-3}m/s.

P30 The variation of atmospheric radioactive ceasium concentration from Fukushima Dai-ichi nuclear accident in Tsukuba and Iitate, and the factors of high concentration phenomena

DOI, T.[1], TAKAGI, M.[1], TANAKA, A.[1], KANNO, M.[2], DOKIYA, Y.[2], TAO, Y.[2], MASUMOTO, K.[3]
([1]National Institute for Environmental Studies, [2] Resurrection of Fukushima, [3] High Energy Accelerator Research Organization)

Radioactive Cs concentration in the atmosphere of Tsukuba and Iitate in about one year after nuclear accident were levels of 10-5 Bq m-3 and 10-4 Bq m-3, respectively. Sometimes atmospheric radioactive Cs at two sites of Iitate showed high concentration due to strong wind and resuspension of soil at the time of local decontamination. The high concentration phenomenon on August 2013 seemed to be caused by the removal of the debris at Fukushima Dai-ichi Nuclear Power Plant. The activity median aerodynamic diameter of 134Cs- and 137Cs-bearing particles were larger than 1.2 µm. Radioactive Cs aerosol of particle size larger than 1 µm may be taken into the alveoli by respiration, but it will be removed thereafter. It was thought that the influence of annual internal radiation exposure by breathing estimated from the atmospheric concentration was low. It was thought that the influence of annual internal radiation exposure from breathing estimated from atmospheric concentration was low.

P31 Formation of groundwater with high radium in the former mining open-pit at Ningyo-toge Environmental Engineering Center of the Japan Atomic Energy Agency

TAKEUCHI, E.[1], TOMITA, J.[1], OHARA, Y.[1]

([1]JAEA)

To investigate the formation of fresh groundwater with high ^{226}Ra activity, groundwater samples discharged from uranium mill tailings, weathered granite and sedimentary rocks were collected from 9 boreholes in a former mining open-pit at Ningyo-toge Environmental Engineering Center, Japan Atomic Energy Agency. Activities of Ra isotopes (^{226}Ra, ^{228}Ra) of groundwater samples were measured along with dissolved cations and anions. Rock samples were also sampled from 8 boreholes, and the activities of U-series (^{238}U, ^{234}U, ^{230}Th, ^{226}Ra) and Th-series (^{232}Th, ^{228}Ra) radionuclides were determined.

The ^{226}Ra activities of the groundwater samples ranged widely from 26 to 2425 mBq kg^{-1}, and the highest value was obtained from the borehole of the uranium mill tailings. The ^{226}Ra concentrations showed an increasing tendency with increase of salinity. In comparison with the groundwater from other areas in Japan, the groundwater samples in this study showed higher ^{226}Ra activities than those estimated from salinities. The results of both ^{228}Ra/^{226}Ra activity ratio of the groundwater and U- and Th-series radionuclide activities of the rock samples suggested the possibility that ^{226}Ra was supplied to groundwater by dissolution of the phase containing high ^{226}Ra activity rather than alpha-recoil process for the groundwater sample with the highest ^{226}Ra activity.

P32 Component Analysis by SEM-EDS on Radioactive Particles Emitted from Unit 1 of FDNPP

ISHII, I.[1], SUEKI, K.[1], MATSUO, K.[1], HASEGAWA, R.[1], SATOU, Y.[2], ADACHI, K.[3], IGARASHI, Y.[4]

([1]Univ. of Tsukuba, [2]JAEA, [3]Mteorological Research Institute, [4]Ibaraki Univ.)

Radioactive particles were released into the environment by the accident of Fukushima Daiichi Nuclear Power Plant (FDNPP). They have information to understand the inside of the reactor during the accident. In this study, we picked up about 50 radioactive particles derived from the Unit 1 and measured them using a Scanning Electron Microscope (SEM) with an Energy Dispersive X-ray Spectrometer (EDS). For each radioactive particle, several point analyses were performed and elemental X-ray intensities were calculated. The X-ray intensities were normalized by that of silicon. Two component plots of the normalized X-ray intensities showed correlations among selected elements. For instance, there was a positive correlation between Na and Mg and also between K and Ca. On the other hand, there was a negative correlation between

the group of Na-Mg and the group of K-Ca. The group of K-Ca also had Ti, Fe, Zn and Pb. We discuss the formation mechanism of radioactive particles with the elemental correlations.

P33 U-236 and uranium isotope ratios in soil samples from Hunan Province, China

SHAO, Y.[1,2], YANG, G.[1], TAZOE, H.[1], YAMADA, M.[1]

([1] Institute of Radiation Emergency Medicine, Hirosaki Univ., Japan, [2] Institute of High Energy Physics, Chinese Academy of Sciences, China)

Without significant nuclear accident, presently, ^{236}U far from the nuclear facilities in China may be mainly from global fallout. In the present study, inductively coupled plasma tandem mass spectrometry (ICP-MS/MS) was applied to analyze ultra-trace ^{236}U in 48 soil samples from Hunan Province, China. The ^{236}U concentrations were measured as $(1.61–21.3)\times10^{7}$ atoms g^{-1}, and the ^{236}U/^{238}U, ^{234}U/^{238}U and ^{235}U/^{238}U atom ratios were $(0.470–4.91)\times10^{-8}$, $(5.10–9.31)\times10^{-5}$ and $(7.11–7.82)\times10^{-3}$, respectively. Comparing with other studies, ^{236}U concentrations in Hunan Province were lower than that in forest soils from Ishikawa Prefectures, Japan ($(8.92–37.6)\times10^{8}$ atoms g^{-1}), similar to soils from La Palma, Spain ($(1.43–8.12)\times10^{7}$ atoms g^{-1}) and soils collected from Fukushima Prefectures, Japan not affected by the NPP accident ($(0.953–20.2)\times10^{7}$ atoms g^{-1}). Considering the facts that neither nuclear tests nor nuclear accidents have occurred in Hunan Province and that the ^{236}U/^{238}U atom ratio results are included in the global fallout value range, the data of ^{236}U in soil samples from Hunan Province may be mainly from global fallout.

P34 Radiochemical analysis of fission and activation products released by Fukushima nuclear accident

KOJIMA, S., ARINOBU, T.

(Aichi Medical Univ., School of Medicine)

We reported previously that the radioactivity of the fission products, ^{127m}Te, ^{129m}Te, ^{125}Sb, ^{137}Cs, ^{140}Ba, ^{141}Ce and ^{144}Ce, and that of the neutron activation products, ^{54}Mn, ^{58}Co, ^{60}Co, ^{110m}Ag, ^{114m}In, ^{134}Cs and ^{136}Cs in the soil sample collected on May 19 at Namie-cho, 8 km northwest from Fukushima Daiichi NPP. The purpose of this paper is to determine the radioactivity of ^{95}Nb and ^{113}Sn in the soil sample. The soil sample was separated into the solution and the insoluble residue by filtration, after it was treated by nitric acid. The gamma-ray of ^{95}Nb was detected in the residue, as Nb hardly dissolved in nitric acid. The radioactivity of ^{95}Nb was 4.9 Bq/g at the sampling time, and it was less than that of one-300th of ^{137}Cs. The gamma-ray of ^{113}Sn was identified in the fraction removed ^{137}Cs and ^{110m}Ag. This means that the atoms of Sn in the cladding tube of zirconium alloy had been irradiated by neutrons, and the produced radionuclides, ^{113}Sn and ^{114m}In, were released from the reactor to the environment.

P36 Study on Migration Behavior of Suspended Solids in the Kumaki River Based on Carbon-Nitrogen Isotope Ratio and Radionuclides

TAHARA, R., NAGAO, S., OCHIAI, S, HASEGAWA, T., GOTO, A.

([1]Grad. School of Natural Sci. and Technol., Kanazawa, Univ., [2]INTE, Kaznazawa, Univ.., [3]Inst. Sci. Eng.LEEE, Kanazawa, Univ..)

Suspended solids in rivers are mainly derived from paddy field and forest soil, industrial and lifework wastewater, resuspended bottom sediments, fallout from the atmosphere. The decline of agriculture and forestry activities has caused the decline of productivity of land and coastal marine environment. In the previous study, ^{7}Be / ^{210}Pb$_{ex}$ ratio is used as an indicator of suspended particle flux due to residence time of recent topography erosion and suspended solids. In this study, we investigated the transport behavior of suspended solids from watershed to river by using the radioactivity(^{7}Be, ^{137}Cs, ^{210}Pb$_{ex}$)and carbon and nitrogen isotopic ratios of organic matter in the Kumaki River located in Noto Peninsula during April 2016 to March 2018. The downward variation of ^{137}Cs/^{210}Pb$_{ex}$ ratio shows difference in every month. However, the ^{137}Cs/^{210}Pb$_{ex}$ ratio of rainy samples shows almost similar values among the up stream, middle stream and downstream.

P37 Composition analysis of radioactive particles composed of two kinds of silicate compounds

MATSUO, K.[1], SUEKI, K.[1], ISHII, T.[1], HASEGAWA, R.[1], SATOU, Y.[2], ADACHI, K.[3], IGARASHI, Y.[4], ABE, Y.[5], NAKAI, I.[5], SAKAMOTO, T.[6], MORITA, M.[6]

([1]Univ. of Tsukuba, [2]JAEA, [3]Meteorological Research Institute, [4]Ibaraki Univ.,
[5]Tokyo Univ. of Science, [6]Kogakuin Univ.)

In March 2011, radioactive particles were emitted into the environment by FDNPP accident. They include much information about the internal reactor building at the time of the accident1). In this study, we found that the radioactive particles from Unit 1 1) were composed of two kinds of silicate materials. Their surface elemental distribution was measured by SEM-EDS and showed that they had Na-abundant part and K-abundant parts. We analyzed them by XRF, XANES and XRD at BL37XU in SPring-8. We found that constituent elements of Na-abundant part differed from these of K-abundant part and that main components of the both part were silicate glass. By using FIB-TOF-SIMS with a fine processing function2), it was revealed that Cs abound in the surface of the particle, and Na-abundant part had more Cs than K-abundant part inside the particle. We also found a clear border between the two parts. From these results, we conclude that the particle was presumed to make by two types of silicate glass and that Cs adhered to the particle after making the matrix of the particle.

1) Y. Satou *et al.* 2018. *Geochem. J.* 52, 137 – 143.

2) T. Sakamoto *et al*. 2008. *Appl. Surf. Sci*. 255, 1617-1620.

P38 Evaluation on Deposition of Fukushima Derived Radiocesium to Bottom Sediment at Mountain Lakes in Gunma Prefecture, Japan

MIYASAKA, S.[1], NAGAO, S.[2], OCHIAI, S.[2], SUZUKI, K.[3], WATANABE, S.[3], ARAI, H.[3], KUGE, T.[3], MORI, M.[4]

([1]Graguate School of Natural Science & Technology, Kanazawa Univ., [2]Institute of Nature and Environment Technology, Kanazawa Univ., [3]Gunma Prefectural Fisheries Experiment Station, [4]Univ. 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 ^{134}Cs and ^{137}Cs was determined by germanium detectors. We also measured the concentration of ^{210}Pb to estimate the sedimentation rate at each sampling site. At Lake Onuma, the ^{134}Cs/^{137}Cs 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 highest concentration of ^{137}Cs for the center of Lake Onuma appeared at 0-2 cm (5.0×10^3 Bq/kg) for the core collected in 2011 and at 5-6 cm (3.7×10^3 Bq/kg) depth in core collected in 2017. The sedimentation rate for the center of Lake Onuma in 2011 (0.060 g/cm^2/y) was similar to that of in 2017 (0.058 g/cm^2/y). Based on above results, sediments associated with ^{137}Cs could deposited from the surface layer (0-2 cm) to deeper layer (5-6 cm) with constant sedimentation rates between 2011 and 2017.

P39 Mössbauer Spectroscopic Characterization of Iron in Iron-loaded Woody Charcoals

SUZUKI, K.[1], SUZUKI, T.[1], YAMAUCHI, S.[2], KURIMOTO, Y.[2], TAKAYAMA, T.[3], SAKAI, Y.[3]

([1]Kitami Inst. of Technology, [2]Akita Prefectural Univ., [3]Daido Univ.)

Woody charcoal is versatile material and has been used as adsorbent due to its porosity. Some metal salts serve as an effective catalyst for carbonizing wood. Among the salts, iron salts show high catalytic activity for wood carbonization. The iron-loaded woody charcoals we synthesized have a mesoporous-rich body and high crystallinity compared to charcoals synthesized without catalysts. The charcoals probably contain magnetic iron species when they are carbonized at relatively high temperature. Thus, it can be expected that the magnetic charcoals adsorb the substances that ordinary woody charcoals are incapable of capturing. We attempted to identify the magnetic iron species included in iron-loaded charcoals by Mössbauer spectroscopy, revealing that the iron species and their production ratio were strongly affected by carbonization method. It is interesting to note that the magnetic iron species produced by a one-step carbonization were extremely different from those by a two-step carbonization.

P40 Feasibility study for identification of radioactive materials sealed in container using negative muon

KUDO, T.[1], NINOMIYA, K.[1], KUBO, K.[2], SHINOHARA, A.[1], TAMPO, M.[3], MIYAKE, Y.[3]

([1]Graduate School of Science, Osaka Univ., [2]College of Liberal Arts, International Christian Univ., [3]Institute of Materials Structure Science, High Energy Accelerator Research Organization)

For identification of radioactive materials, many analysis methods have been applied such as ICP-MS, fluorescence X-ray analysis and radioactivity measurement. Among them, radioactivity measurement is the most powerful tool because this method can be applied without sample destruction if the sample is sealed in the container. However, it is difficult to apply radioactivity measurement for the sample with low gamma-ray branching nuclide and/or existing strong gamma-ray emission impurities.

Our group has been studying on development of elemental analysis methods using muonic X-ray. Muonic X-rays emitted from muonic atom has 207 times higher energy than these of electronic X-rays. In this study, we performed muon irradiation experiment for bismuth metal sealed in the container and examined feasibility of analysis by muonic X-rays for such samples. The muon irradiation experiment was performed at D2 beam line in J-PARC/MLF, world's intense pulsed muon source. Muonic X-rays were measured by germanium detectors.

By selecting the timing of muon beam pulse, background signals were significantly reduced, and we successfully identified Bi sealed in Aluminum container without sample destruction.

P41 Local Fields in ZnO Codoped with Co and Mn —a ^{57}Co Emission Mössbauer Spectroscopic Study—

MORIICHI, R.[1], KANO, Y.[1], SATO, W.[1, 2]

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

We measured internal fields in 6.25 at.% Co- and 6.25 at.% Mn-codoped ZnO (CMZO) and 11 at.% Co-doped ZnO (CZO) synthesized by a solid state reaction by means of ^{57}Co emission Mössbauer spectroscopy. In CMZO, two doublets were observed in the spectrum at room temperature : one (D1) is assignable to a $^{57}Fe^{2+}$ ($\longleftarrow ^{57}$Co) substituting for a defect-free Zn site in ZnO, and the other to $^{57}Fe^{3+}$ ($\longleftarrow ^{57}$Co) in substitutional Zn sites adjacent to a Zn vacancy. However, below 100 K, the spectrum cannot be fitted only with two doublets, so we assumed the emergence of an additional magnetic component (M1). We found that the relative intensity of D1 decreases and that of M1 increases with decreasing temperature. In addition, we carried out magnetization measurements for CMZO, CZO and 6 at.% Mn-doped ZnO (MZO), and found that only CMZO shows slightly different trend from that expected for paramagnetism. The results of the ^{57}Co emission Mössbauer spectroscopy and magnetization measurements may imply that codoping Co and Mn in ZnO can realize the emergence of dilute magnetism.

P42 Effect of molecular structure on the muon transfer process in benzene and cyclohexane

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

([1]Grad. Sch. Sci., Osaka Univ., [2]RCNP, Osaka Univ.)

Formation processes of muonic atoms are influenced by the chemical environments of muon-capturing atoms, as it is called chemical effect. Previously, we reported that the chemical effect on the muon transfer processes in benzene and cyclohexane in the gas phase was smaller than the detection limit. In this study, to investigate the chemical effect on the muon transfer processes in the liquid phase, muon irradiation experiments for liquid mixtures of benzene or cyclohexane and carbon tetrachloride were carried out. The experiments were conducted at the MuSIC-M1 beamline of the Research Center for Nuclear Physics (RCNP) in Osaka University. Muonic X-rays emitted after muon-capture by carbon or chlorine atoms were measured with high purity germanium detectors. The muon capture probabilities of carbon and chlorine atoms were determined from the muonic X-ray spectra. The model formula for the muon capture was set up, and the muon transfer rate to carbon atoms in each sample was obtained using the muon capture probabilities. The result suggested that the muon transfer rate to carbon atoms of benzene molecules are higher than that to carbon atoms of cyclohexane molecules.

P43 Observation of local structures in perovskite oxide $SrTiO_3$ with the ^{111}Cd probe

KOMATSUDA, S.[1], SATO, W.[2], OHKUBO, Y[3].

([1]Nat. Inst. Tech., Ichinoseki College, [2]Inst. Sci. Eng., Kanazawa Univ., [3]KURNS)

Strontium titanate ($SrTiO_3$) doped with metal ions as impurity donors exhibits a wide variety of electric properties. For a practical use of $SrTiO_3$ as a conduction-controlling device, it is of great importance to study the lattice site location of doped impurities and their surroundings. For that purpose, the time-differential perturbed angular correlation method (TDPAC) is very suited because it can directly provide atomic-level information of impurity atoms. From this point of view, we have applied the TDPAC method to a study of $SrTiO_3$ using the ^{111}In(→^{111}Cd) probe. Strontium titanate sample doped with the ^{111}In(→^{111}Cd) was synthesized by a solid-state reaction in air at 1473 K. For the PAC spectrum obtained at room temperature, two electric field gradients were observed, characterized by the quadrupole frequencies and its fractions ω_{Q1} = 48.9(1) Mrad/s, f_1 = 68(3)%, and ω_{Q2} = 52.1(2) Mrad/s, f_2 = 36(3)%, respectively. In the presentation, based on the further investigation of TDPAC spectra for $SrTiO_3$ measured at 500~1000 K, we discuss the site occupation of the ^{111}In(→^{111}Cd) probe and their local structures in perovskite oxide $SrTiO_3$.

P44 Hydrogen-Induced Defects in Palladium Observed by Means of Gamma-Ray Nuclear Spectroscopies

FURUMOTO, M.[1], SHIMIZU, H.[1], OHKUBO, Y.[2], SATO, W.[1,3]

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

Palladium (Pd) can dissolve a large amount of hydrogen atoms in the lattice interstitial sites and is expected to be applied as a hydrogen storage material. However, it is widely known that defects formation induced by hydrogen absorption may decrease its storage performance. Thus, application of Pd to hydrogen storage metal still has problems in terms of tolerance. Therefore, in order to obtain more detailed information on the hydrogen storage performance of Pd, it is important to investigate the physical state of lattice defects caused by the introduction of hydrogen. In this study, behavior of lattice defects in Pd associated with hydrogen absorption was observed by two different nuclear spectroscopies. Positron annihilation lifetime spectroscopy (PALS) applied to electrochemically hydrogen-introduced Pd suggested existence of lattice defects. Perturbed angler correlation (PAC) measurements using the ^{111}Ag probe revealed the presence of electric field gradient at the probe site in the electrochemically hydrogen-introduced Pd. In the poster presentation, we evaluate the hydrogen-induced lattice defects in Pd based on the results of the nuclear spectroscopic methods.

P45 Production method development of ^{67}Cu using thermoseparation for cancer therapy.

OHTA, A.[1,2], KAWABATA, M.[1,2], MOTOISHI, S.[1,2], SAEKI, H.[1,2], HASHIMOTO, K[2]., TSUKADA, K.[2,3], HATSUKAWA, Y.[2], NAGAI, Y.[2]

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

^{67}Cu is a promising RI for both cancer therapy and diagnosis in nuclear medicine, yet the large scale production has not been achieved to satisfy the clinical demand. High quality ^{67}Cu can be produced via the ^{68}Zn(n,x)^{67}Cu reaction（x=n'p, d）from zinc target where projecting fast neutrons are generated in carbon converter by stopping 40 MeV deuterons from an accelerator. Separation of ^{67}Cu from ^{68}Zn were previously established1), however it may require a longer hours to obtain a final product from a large volume of zinc target. Concentration of a large quantity of zinc was therefore investigated by thermoseparation2) using a difference in vapor pressure between zinc and copper. The method also serves the removal any impurity copper in the zinc target before irradiation.

Natural zinc metal that was irradiated by neutrons at CYRIC Accelerator (Tohoku University) or Tandem Accelerator (Japan Atomic Energy Agency) was heated under reduced pressure. The bulk of zinc was sublimated, moved and deposited at lower temperature area. The gamma-rays from ^{67}Cu and ^{69m}Zn within the target were measured with CZT (CdZnTe) semiconductor detector. The poster discusses the results on separation efficiencies and the behavior of both tracer elements while they were heated.

Reference

1) Y. Sugo et. al., J. Phys. Soc. Jpn. 86, 023201 (2017)
2) Lu Zhan et. al., Separation and Purification Technology 68, 397 (2009)

P46 Development of a Rn-At generator system using radon gas enclosed within a syringe

KAWASAKI, K.[1], SHIN, Y.[1], AOI, K.[2], WASHIYAMA, K.[3], NISHINAKA, I.[4], HABA, H.[5] YANOU, S.[5] YOKOYAMA, A[2]

([1]Graduate School of Natural Science and Technology, Kanazawa University., [2]Institute and College of Science and Engineering, Kanazawa University., [3]Fukushima Medical University., [4]Tokai Quantum Beam Science Center, QST., [5]RIKEN Nishina Center.)

The radionuclide of ^{211}At gathers much attention as a nuclide useful for isotope treatment. Its chemical species are explored and the solvent extraction of At is investigated for that purpose. However, its half-life is as short as 7.2 hours and it is difficult to use it in a place remote from an accelerator. Milking of ^{211}Rn (of 14.6 hours in half-life), the parent nuclide of ^{211}At, makes it possible to use it at a remote location. In this study, ^{211}Rn gas was stored in a sealed syringe form a dissolved target. ^{211}At produced ^{211}Rn gas with nitric acid or hydrochloric acid. Beside we investigated on the extraction and back extraction of ^{211}At using ionic liquid as solvent and crown ether as extractant. As a result, we succeeded in holding ^{211}Rn gas in a syringe and recovering ^{211}At from ^{211}Rn gas. The ^{211}At extraction was performed at an extraction rate of about 90％ with 0.1M nitric acid. The back extraction of At was performed with sodium hydroxide and potassium hydroxide at their higher concentrations than 1M. From the results, we conclude that this method is promising as a generator system.

P47 Syntheses and stability evaluation of alkaline earth metal complexes for nuclear medicine

NAGATA, K.[1], YAJIMA, T.[2], KASAMATSU, Y.[3], SHINOHARA, A.[3], YOSHIMURA, T.[1]

（[1]Institute for Radiation sciences, Osaka Univ., [2]Materials and Bioengineering, Kansai Univ., [3]Graduate School of science, Osaka Univ.）

Radium-223(^{223}Ra) has a great potential for targeted alpha-ray therapy due to multiple alpha decay in the decay chain. The maximum value of the stability constants $\log K_{\mathrm{ML}}$ reported in the known Ra(II) complexes still remains at ~10, and the $\log K_{\mathrm{ML}}$ value is insufficient to apply in nuclear therapy. We have tried to synthesize a stable Ra complex *in vivo* to obtain new cancer drugs applicable to various kinds of tumors. In the present study, we used macrocyclic ligands, **L1** and **L2**, that have two 6-methyl picolinate arms and a diaza-18-crown-6-ether framework.

The alkaline earth metal (Ca, Sr, Ba) complexes were synthesized by the reactions of **L1** and **L2** with the metal ions, respectively. The stability constants in the Ca(II), Sr(II), and Ba(II) complexes with **L1** and **L2** were determined by pH-potentiometric titration in 0.1 M KNO_3(aq). In all the metal ions studied, the $\log K_{\mathrm{ML}}$ values in the **L1** complexes are high compared with the values in the **L2** complexes. The $\log K_{\mathrm{ML}}$ values follow the order: Ba(II) > Sr(II) > Ca(II) for both the **L1** and **L2** complexes. It has been found that **L1** can provide radionuclide (^{85}Sr, ^{133}Ba and ^{226}Ra)-chelate complexes. The reactions detected by the radio TLC method with γray spectroscopy. The complex formation was quantitative in both the cases using ^{85}Sr, ^{133}Ba and ^{226}Ra.

P48 Production of ^{211}At using middle-sized cyclotron, MP-30 and its purification

WASHIYAMA, K.[1], AOKI, M.[2], SASAKI, S.[2], ODA, T.[3], NISHIJIMA, K.[3], TAKAHASHI, K.[1]

([1]Advanced Clinical Research Center, Fukushima Medical Univ., [2]SHI Accelerator Service Ltd., [3]Sumitomo Heavy Industries, Ltd.)

Among the α-emitters that can be used for targeted radionuclide therapy, Astatine-211 with a half-life of 7.21 hours is one of the promising isotopes for clinical application. There have been many institutes and universities reporting to study ^{211}At for production, labeling, preclinical evaluation in Japan. Fukushima Medical University has also started production of ^{211}At using the medium-sized cyclotron MP-30 and its vertical irradiation system developed by Sumitomo Heavy Industries, Ltd. since 2016. We irradiate Bi-metal with 28.6 MeV α beam of 20 eμA to produce ^{211}At by 209Bi(α, 2n)^{211}At nuclear reaction. The thick target yield is 30.2 MBq/μA·h as almost same as in the literature. Here we report our evaluation regarding the series of changes the beam current, energy, Bi-metal amount, target holder design, to find the optimum conditions of our targetry.

P50 Investigation on solvent extraction of astatine for ^{211}Rn-^{211}At generator

—Comparison to the ^{131}I behavior and investigation on the effect of oxidizing agents in extraction

SHIN, Y[1]., KAWASAKI, K[1]., AOI, K[2]., YOKOYAMA, A[2]., WASHIYAMA, K[3]., NISHINAKA, I[4]., YANOU, S[5]., HABA, H[5].

([1]Graduate School of Natural Science and Technology, Kanazawa Univ., [2]Institute and College of Science and Engineering, Kanazawa Univ., [3]Fukushima Medical Univ., [4]Tokai Quantum Beam Science Center, QST., [5]RIKEN Nishina Center.)

^{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 are developing a ^{211}Rn-^{211}At generator. In a previous study, ^{211}At produced in decaying of ^{211}Rn in dodecane solution can be back-extracted into 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 from NBS oxidizes astatine. We used NCS and NIS instead of NBS to investigate the effect of redox potential. In addition, we compared the back-extraction ratios of ^{211}At and ^{131}I by using bromine water as an oxidizing agent. As a result, the order of back extraction ratio is NBS > NCS > NIS. Comparing ^{211}At and ^{131}I, the back-extraction ratio of ^{211}At increases with addition of bromine, but that of ^{131}I does not increase. It shows that ^{211}At is oxidized into species which is easily extracted.

P51 Synthesis of an astatinated amino acid derivatives via silicon-astatine exchange reaction

WATANABE, S.[1], ANWAR-UL, A.M.[1,2], NISHINAKA, I.[1], SASAKI, I.[1,3], OHSHIMA, Y.[1], YAMADA K.[3], ISHIOKA, N.S.[1]

([1]QST Takasaki, [2]NINMAS, BAEC, [3]Graduate School of Science and Technology, Gunma Univ.)

Astatine-211 (^{211}At) is a promising alpha-emitting radionuclide for the targeted alpha therapy (TAT) and tin-astatine exchange reaction has been used for the synthesis of astatinated compounds. However, organotin compound is readily decomposed under high temperature and acidic condition, which makes synthetic routes of precursor complicated. In addition, toxicity of the organotin compounds is a potential cause of concern for future clinical applications. We therefore focused on silicon-astatine exchange reaction. In the present study, 4-[^{211}At]astato-L-phenylalanine was synthesized via the electrophilic desilylation. HPLC analysis revealed that a strong radioactive peak was observed at 16.8 min. of retention time, which was identical to that of corresponding radioiodinated phenylalanine (retention time: 15.8 min.). The astatinated compound was time-dependently accumulated into LS180, colon adenocarcinoma cell, and the uptake was inhibited in the presence of some amino acid derivatives. These results clearly indicated that 4-[^{211}At]astato-L-phenylalanine was successfully synthesized in this study. Radiochemical yield and purity were 64-75% and over 99%, respectively. We therefore make a conclusion that the silicon-astatine exchange reaction is useful to synthesize astatinated amino acid derivatives and peptides.

P52 Study on synthesis of Y@C_{60} by nuclear recoil

ITO, Y.[1],AKIYAMA, K[1], KUBUKI, S.[1], HATSUKAWA, Y.[2]

([1]Tokyo Metropolitan Univ, [2]QST.)

We have tried to synthesis of Radioactive Endohedral Metallofullerenes (REMFs) using the nuclear recoil induced by nuclear reaction. This method has some advantages from the viewpoint of a small amount of by-products and selective production of target REMFs. In the previous study, we reported the synthesis of ^{85}Sr@C_{60} using the nuclear recoil ^{86}Sr(n, 2n)^{85}Sr reaction with 14 MeV fast neutron induced by DT reaction. In this study, we will report the synthesis of ^{88}Y@C_{60} by nuclear recoil of ^{89}Y(n, 2n)^{88}Y reaction with 30 MeV and 50 MeV fast neutron induced by DT reaction.

Irradiated samples were dissolved into CS_2 and filtered to remove a large amount of C_{60}. After dryness of the filter, aniline and HCl_{aq} was passed through the filter, sequentially. The γ ray from extracted samples was measured by Ge detector.

As a result, the radioactivity of ^{88}Y observed in the aniline solutions derived from ^{88}Y@C$_{60}$ for examined in this work was estimated to be several percents in the total radioactivity of each irradiated sample.

P53 Preparation of Carrier-free ^{139}Ce Tracer for Endohedral Metallofullerene Study

AMEKURA, K. [1], AKIYAMA, K. [1, 2], KUBUKI, S. [1], HABA, H [2]

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

We have studied the electronic state for a series of light lanthanide (Ln) metallofullerene, such as Ln@C_{82}, by analyzing the small difference in the HPLC retention time. For such precise analysis, the radiochemical method is one of the most effective approach because this method allows the simultaneous analysis of multiple elements. For further studies, it is necessary to analyze from the difference between these retention times monitored by gamma ray of reference carrier-free Ln@C_{82} and by UV absorption because some heavy lanthanide, such as Er and Dy, only produce nuclides with short half-life. In this study, we report the chemical separation of ^{139}Ce used as a carrier-free reference material from a target material of La and the result of metallofullerene preparation using this carrier-free ^{139}Ce.

^{139}Ce and a target material of La were separated by the solvent extraction method using $K_2Cr_2O_7$ as an oxidant. The amount of ^{139}Ce was determined by gamma-ray measurement and the residual amount of Cr and La were estimated to be 2.5% and 0.03%, respectively, from UV/vis absorption analysis. More than 90% of ^{139}Ce were finally obtained and also EMF is found to be successfully prepared using this carrier-free ^{139}Ce.

P54 A study on the sedimentary environment of Tokyo-bay sediments under hypoxia.

MATSUO, M.[1], KOMIRI, M.[1,2], SHOZUGAWA, K.[1], OKUMURA, R.[3], IINUMA, Y.[3], TAKAMIYA, K.[3]

([1]Graduate School of Arts and Sciences, The University of Tokyo, [2]Yokohama Environmental Science Research Institute, [3]KURNS)

Hypoxia is a big problem particularly in inner bay such as Tokyo Bay. To understand and solve the problem, we attempted to estimate the redox environment of Tokyo-bay sediments under hypoxia, assuming that the effect of past hypoxia could be preserved as a difference in concentrations and/or chemical states of elements in the sediments.

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 by instrumental neutron activation analysis.

As a result, the concentration of Mn was low in the dredged trench off Makuhari, whereas there was no significant difference in Fe. It is estimated that the oxidation-reduction potential in the dredged trench remained to the extent that Fe precipitation occurred but no Mn precipitation occurred. In addition, the values of Th/U and Ce/U ratios at each site existed in different places on a same line. It is found that the magnitude of the reductive environment increased in the order of flat seafloor off Yokohama < flat seafloor off Makuhari < dreaded trench off Makuhari. This trend is in good agreement with water quality data and estimation results by ^{57}Fe Mössbauer spectroscopy.

P56 Production and Purification of ^{105}Ag

KUBOTA, T.[1], IWATA, K.[2], FUKUTANI, S.[1], TAKAHASHI, T.[1], TAKAHASHI. S.[1]
([1]KURNS, [2]Graduate School of Agriculture, Kyoto University)

The Fukushima nuclear accident released a large amount of radioactive nuclides into the environment. Among them, short half-life radionuclides, for example ^{110m}Ag, would be paid less attention to; however, those are critical for external and internal exposure just after the accident. In this study we prepare carrier-free radioactive silver, ^{105}Ag, as tracer nuclide, which is suitable for plant and animal experiment. ^{105}Ag was produced from cadmium through photo-nuclear reaction. An aliquot of $CdCl_2$ of 2.5 g in a quartz test tube was dehydrated in a muffle furnace and encapsulated under vacuum. The test tube was irradiated with photons generated by the bombardment of platinum sheets with 30 MeV-electrons. ^{105}Ag was purified by precipitation and anion exchange chromatography. Cadmium was removed by precipitation at pH 11 in chloride solution, while silver remained in the solution as complex ion. Cadmium and silver was once adsorbed onto anion exchange resin, then cadmium was washed out with 0.1 M HCl and finally silver was recovered with 0.1 M HNO_3. The resulting product showed that the specific activity of ^{105}Ag was 6.8 (kBq/mL) and the concentration of cadmium was 10 ppb.

P57 Non-destructive elemental analysis of Ni based super alloys by using time of flight prompt gamma-ray analysis

TOH, Y. [1], HUANG, M.[1,2], SEGAWA, M.[1], MAEDA, M.[1], TSUNEYAMA, M.[1,3], KIMURA, A.[1], NAKAMURA, S.[1]
([1]JAEA, [2]IMP, [3]NAT.)

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). We have developed an analytical technique which combines prompt gamma-ray analysis with the time-of-flight technique by using an intense pulsed neutron beam. We have conducted several experiments of Ni-based super alloys samples, and have analyzed 3 types of spectra PGA, TOF(NRCA) and TOF-PGA. Presentation will describe the detail of the experimental results and new device (neutron filter), the current status of the developments and future prospects.

This work was supported in part by Grants-in-Aid for Scientific Research (JP17H01076).

P58 LET Dependence of Reactive Oxygen Species Generations in an Aqueous Sample Irradiated by Heavy-Ion Beam

MATSUMOTO, K., NYUI, M., UENO, M., OGAWA, Y., NAKANISHI, I.

(Quantitative RedOx Sensing Team, Department of Basic Medical Sciences for Radiation Damages, National Institute of Radiological Sciences, National Institutes for Quantum and Radiological Science and Technology)

The amounts of reactive oxygen species generated in aqueous samples by irradiation with X-ray or clinical carbon-ion beams were quantified. Hydroxyl radical (•OH), hydrogen peroxide (H_2O_2), and the total amount of oxidation reactions were measured by electron paramagnetic resonance-based methods. •OH generation was expected to be localized on the track/range of the carbon-ion beam/X-ray, and mM and M levels of •OH generation were observed. Total •OH generation levels were identical at the same dose irrespective of whether X-ray or carbon-ion beam irradiation was used, and were around 1.2–1.3 μmol/L/Gy. However, sparse •OH generation levels decreased with increasing linear energy transfer, and were 0.61, 0.53, and 0.38 μmol/L/Gy for X-ray, 20 keV/μm carbon-ion beam, and 80 keV/μm carbon-ion beam sources, respectively. H_2O_2 generation estimated by TEMPOL decay was as 0.21, 0.57, and 0.35 μmol/L/Gy, for X-ray, 20 keV/μm carbon-ion beam, and 80 keV/μm carbon-ion beam sources, respectively, whereas the H_2O_2 generation estimated by Fenton-reaction-ability was as were 0.22, 0.26 and 0.29 μmol/L/Gy, respectively. TEMPOL decay may only reflect the dense H_2O_2 generation. The amounts of total oxidation reactions were 2.74, 1.17, and 0.66 μmol/L/Gy, respectively. The amounts of oxidation reaction caused by the low LET beam were reduced in hypoxic atmosphere, but not changed by high LET (>80 keV/μm) beam. The generation of reactive oxygen species was not uniform at the molecular level.

P59 Radiocarbon dating of the ancient calligraphy fragments attributed to famous persons

ODA, H.[1], IKEDA, K.[2], YASU, H.[3], SAKAMOTO, S.[4]

([1]ISEE, Nagoya Univ., [2]Faculty of Letters, Chuo Univ., [3]Taga High School, [4]Ryukoku Univ.)

Kohitsugire are ancient paper sheets containing elegant calligraphy. They were originally pages or parts of ancient manuscripts, pictures or sutras. The old calligraphy written before the 14th century hardly remain as complete books or scrolls; therefore, *kohitsugire* potentially has high academic value. Most of *kohitsugire* have traditional certificates which identify the calligraphists. However, recent paleographical study shows that some of them are not genuine handwriting. Therefore, we applied radiocarbon dating to such *kohitsugire* attributed to famous persons: Ono no Tohu (894-966), Saigyo

(1118-1190) and Retired Emperor Gotoba (1180-1239). The radiocarbon dating indicated that all of them were not their genuine handwritings. They were written by different persons in a few century later. It suggested that many copies and counterfeits are in circulation among *kohitsugire* attributed to famous persons.

P60 Difference in electrolytic deposition of uranium and neptunium

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

([1] Japan Atomic Energy Agency)

In the previous studies, we focused on deposition following valence changes and investigated the electrolytic reduction of U(VI) in weak acid solutions. We found that U(IV) deposits form on the electrode surface by the electrolytic reduction of U(VI) to U(V). Also, we proposed the deposition mechanism that a U(IV) hydroxide formed as an intermediate and transforms to U(IV) oxide as a terminal product. In this study, we investigated the electrolytic deposition of Np(V) by measuring electrochemical quartz crystal microbalance (EQCM) to understand the difference in electrolytic deposition among actinides. The changes of the reduction current and deposition amount with time during -0.75 V was applied to reduce Np(V) were measured. The reaction stopped when the electrode surface was covered with Np deposits. This behavior is different from the deposition of U that the reaction continues even if deposits form on the electrode surface. From the relationship between the quantity of electricity of electrolytic reduction of Np(V) and the weight of deposits in the pH 4 solution, we found that the specie of deposits is highly likely to be Np(IV) dioxide (NpO_2) and the NpO_2 on the electrode surface prevent from reducing Np(V) to Np(IV) because of high electrical resistance of NpO_2.

P61 Safety verification by experimental data for rational regulation of short-lived α-ray emitting nuclides and development of radiation safety management and educational method

SHINOHARA, A.[1,2], YOSHIMURA, T.[3], TOYOSHIMA, A.[2], KANEDA, K.[1,2], ZHANG, Z.[1], NAGATA, K.[3], WATABE, T.[4], OOE, K.[4], HATAZAWA, J.[4], YAMAMURA, T.[5], SHIRASAKI, K.[6], KIKUNAGA, H.[7], HABA, H.[8], WASHIYAMA, K.[9]

([1]Graduate School of Science, Osaka Univ., [2]Institute for Radiation Sciences, Osaka Univ., [3]Radioisotope Research Center, Osaka Univ.. [4]Graduate School of Medicine, Osaka Univ., [5]IRNS, Kyoto Univ., [6]IMR, Tohoku Univ., [7]ELPH, Tohoku Univ., [8]RIKEN, [9]Fukushima Med. Univ.)

Research on short-lived alpha-particle emitting nuclides for medical applications is underway extensively. It is expected that large scale production and medical use of these nuclides will become popular in the near future. To ensure reasonable radiation management to support the research and development while ensuring workers and public safety, it is essential to acquire experimental data on diffusion rate, exhaustion, waste water and surface pollution, which are the basis of laws and safety management, and to develop a management method. In this study, some experiments as models for RI production, purification, labeling (chemical synthesis), and animal experiments were conducted for the purpose of acquiring the data of ^{211}At, ^{223}Ra and ^{225}Ac and their disintegrated nuclides. In addition, it was done to investigate the working situation of radiation workers and medical workers, and to prepare an effective education and training program.

P62 An attempt to estimate ^{111}In activity by the sum-peak method

OGATA, Y.[1], KOJIMA, S.[2]

([1] RIC Medical Division, Nagoya Univ., [2]Aichi Medical Univ.,) 200 words

Indium-111 is used for the bone marrow scintigraphy in nuclear medicine. It decays with a half-live of 2.8 days by electron capture mainly emitting 171 keV and 245 keV cascading gamma-rays. We tried to determine the radioactivity by the sum-peak method and the modified sum-peak method. A radiopharmaceutical injection solution of ^{111}InCl, 74 MBq mL^{-1}, was diluted and about 50 kBq of the solution was deposited on a thin grass filter and dried. The source was measured by a HPGe detector. The source was put on an axis of the detector varying the source-to-detector distance. The calculated activities were 67 MBq and 70 MBq by the sum-peak method and the modified sum-peak method, respectively. Although, the source actuary included ^{114m}In, it did not affect the calculation. It was confirmed that the activity of ^{111}In can be determined by the sum-peak method.

P64 Non-destructive elemental analysis by using characteristic muonic X-rays

KURODA, A.[1], TAMPO, M.[2], NINOMIYA, K.[3], TAKESHITA, S.[2], MIYAKE, Y.[2], SAITO, T.[4] , KUDO, T.[3] , FUJIO, S.[4], UENO, Y[4], KUBO, M. K.[1]

([1]Internatinal Christian University, [2]J-PARC, [3]Graduate School of Science, Osaka University, [4]National Museum of Japanese History)

A bronze mirror made in China in 2nd century was analysed using negative muon beam at D2 port in J-PARC MLF in this experiment. The diameter of the mirror was about 12 cm and 6 positions on the mirror side were measured. Making process of bronze mirrors is yet unknown in detail. However, by using negative muon beam, non-destructive elemental analysis becomes possible with selectivity in depth. This method allows us to ignore the surface rust and analyze the elemental composition of interior part. We used 4 germanium detectors and placed them around the sample. Each position is irradiated for about 10 hours to the negative muon beam. The intensity of the proton beam was 500 kW and the momentum of the muon bean was 35 MeV, which makes muon to stop about 400 μm from the surface of the sample.

From the spectrum of muonic characteristic X-rays, peaks of Cu, Sn, and Pb are clearly observed. Since this method using characteristic muonic X-rays is sensitive to all of the existing elements in the sample, we confirmed that this bronze mirror consists of Cu, Sn and Pb.

P65 Actinide Molecular Ion Formation in Collision/Reaction Cell of Triple Quadrupole ICP-MS/MS

SUZUKI, T.[1], YAMAMURA, T.[2], ABE, C.[3], KONASHI, K.[3], SHIKAMORI, Y.[4]

([1]Nagaoka Univ. Tech., [2]IIRNS, Kyoto Univ., [3]IMR, Tohoku Univ., [4]Agilent Technologies Japan)

Recently, the actinide analysis by using ICP-MS has been developing rapidly. In the present work, a fundamental studies on actinide analysis by quadrupole ICP-MS/MS, Agilent 8900, were carried out. The used nuclides were U-238, Np-237, Pu-240, Am-241, and Cm-244, respectively. Np-237 and Pu-240 were used the daughters of Am-244 and Cm-244, respectively. We confirmed the formation percentages of ionic species of each actinides in several kinds of collision/reaction cell modes; no gas, hydrogen gas, helium gas, oxygen gas, and BEC (background equivalent to counts) and LOD (limitation of detection) of each actinide ion are confirmed in each mode. We also investigated the formation percentages of molecular ions (monoxide ions and dioxide ions) in the case of oxygen gas mode. We found that the formation percentages of actinide ion, monoxide ion, and dioxide ion depends on the oxygen flow rate and on actinides; the dioxide ion of actinide with lower atomic number is easily formed. These phenomena can be explained from a view point of electron configuration of actinides. The obtained results indicate the feasibility of discrimination of isobaric interference ions.

P66 Gamma-ray irradiation impact of humic substances on apparent formation constants with Cu(II)

SASAKI, T.[1], GOTO, R.[1], SAITO, T.[2], KOBAYASHI, T.[1], SUGIYAMA, Y.[3]

([1]Graduate School of Engineering, Kyoto Univ., [2]KURNS, [3]Department of Applied Science, Okayama Univ. of Science)

Three humic substances (HSs) in solution were irradiated with ^{60}Co gamma-rays in the dose range of 0–500 kGy at two dose rates, 0.1 and 1 kGy/h. The brownish solutions discolored with 500 kGy irradiation due to the degradation by radiolysis, and the index of degree of humification as the absorbance ratio of 400 nm / 600 nm was studied using UV-Vis spectroscopy. Apparent formation constants of Cu(II) with HSs in 0.1 M $NaClO_4$ were determined at 298 K by potentiometric titration using commercially available Cu ion selective electrode, and the effect of gamma irradiation on the constants was discussed. The acid dissociation constants of HSs were also measured in order to determine formation constants by potentiometric titration. No significant change in apparent formation constants was found, though the concentration of carboxylic groups decreased drastically with high radiation exposure.