English abstracts

2S01 Establishment and the present status of animal archives in and around the ex-evacuation zone of the Fukushima Daiichi Nuclear Power Plant (FNPP) accident

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The Fukushima Daiichi Nuclear Power Plant (FNPP) accident released large amounts of radioactive substances into the environment. Since the FNPP accident, major concern in the world has been the health effect of long-term low dose exposure to both internal and external radiations. However, it is impossible to perform such exposure experiments including laboratory animals. The very important characteristics of the FNPP accident is that the accident happened in Japan which is one of the most advanced and open countries and the amount of released radionuclides was 1/10 of the Chernobyl accident. We collected organ specimens from cattle, pigs and wild Japanese monkeys within and adjacent to the 20-km radius previously set as the evacuation zone of FNPP. We have been performing determination of organ concentrations of radionuclides and their biological effects. I will address how we have established the archive system, what we knew using our archive system, and problems we are facing at.

1A01 Differences by the source reactor of the radioactive particles emitted from the Fukushima Dai-ichi Nuclear Power Plant accident

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The radioactive particles have been emitted to environment in the Fukushima Daiichi Nuclear Power Plant accident in 2011. At least these particles were emitted from the units 1 and 2. However, the details have not been clarified yet. This study performed isolation of radioactive particles including so-called Cs bearing particles, from soil samples, and performed characterization. And we disclosed differences between the particles from the units 1 and 2. We isolated each four radioactive particles from two soil samples. The radionuclides ^{134}Cs and ^{137}Cs were detected by gamma spectroscopy in all of the particles. However, no other gamma emitters were detected in the particles. The specific radioactivity of particles from unit 1 is lower than that from unit 2. A significant amount of Ba has been detected in the particles from unit 1. However, distribution of Ba were different from that of Cs. For this reason, origin of Ba could not be a decay product of Cs. In addition, a distribution of Fe and Zn were not consistent with that of Cs. In contrast, Si and Ca were detected in common. Si is one of major components in the radioactive particles emitted by the Fukushima accident.

1A02 Development of rapid separation method of plutonium and americium contained in the soil sample

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Artificial elements of Pu and Am generated in nuclear fuel are known to be released from Fukushima Nuclear Power Plant in March, 2011. In this study, we aim to develop a rapid quantitative analysis of Pu and Am in the soil sample for nuclear emergency. We focused firstly on dissolution and pre-concentration, and secondly, on speeding up purification for Am in the sequential analysis. The ammonium nitrate fusion and the purification of Am by a DGA resin column have been examined. In the procedure, DDTC solvent extraction was found necessary to eliminate polonium atoms. Finally, we ensured that these procedures were applicable to the soil samples for monitoring contamination in Fukushima.

1A03 A Cesium detachment from the clay minerals using molten salt treatment and structure analysis

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There is the issue of soil contamination problem with the radioactive cesium (Cs) large quantities scattering with the Fukushima first Nuclear Power Plant accident without the still radical solution that nearly four years passed being done by outbreak. The molten salt treatment method is one of the effective Cs removals, however, waste increases in thing and detachment needing a high temperature processing process and still has a problem for practical use. In this study, we investigated the Cs detachment by the low pressure sublimation method using the molten salt (NaCl, $CaCl_2$ mixed salt) and the structure analysis at the Cs removal from clay minerals and a reuse materials search. When we gave low-pressure processing with NaCl, $CaCl_2$ mixed salt and heated, we achieved Cs removal rate 100% at 700 degrees. As a result of structure analysis of the clay minerals which occurred in a heating process using X-rays diffraction method (XRD) and transmission electron microscope (TEM-EDS), Cs formed the plural structure called the Calcite, Hematite, Augite, and Wadalight and knew that Cs was removed while the frame structure of clay minerals changed.

1A04 Dynamism of radioactive cesium in blood and muscle of cattle fed with radioactive contaminated silage

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We investigated the correlation of internal radiocesium deposition determined by blood monitoring and by surface monitoring on the neck or ramp muscle of live-cattle using a NaI surveymeter. Blood and muscle radiocesium concentration was measured in a Ge spectrometer. Seven-month-old or matured Japanese black cattle were fed with radiocesium-contaminated silage for approximately 60 days, thereafter, with contamination free clean silage.

Surface monitoring values (cps) were well correlated with radiocesium concentration (Bq/kg) in the dissected muscle. Therefore, we could convert the surface monitoring value to radiocesium concentration in the muscle. We further compared surface monitoring data with blood monitoring data. Radiocesium appeared to accumulate in muscle and blood with similar kinetics over the period of feeding with the contaminated silage. On the other hand, the decrease in the muscle was slower than that in blood after switching to the clean silage. It was confirmed that biological halflife (T_b) was approximately 2 weeks and 1 month in blood and the muscle, respectively. We also found the age-dependent elongation of T_b in the muscle.

The present study revealed that internal radiocesium deposition can be estimated by surface monitoring, as the noninvasive method.

We will discuss conditions to be considered for calculating surface monitoring values to muscle radiocesium concentrations.

1A05 Strontium-90 in fishes after the Fukushima Dai-ichi Nuclear Power Plant accident

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Radioactive wastewater released from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) includes the high concentration of ^{90}Sr. After the wastewater was discharged into the sea, ^{90}Sr in seawater were only temporarily detected near the FDNPP harbor. Strontium-90 in seawater was spread across the ocean and diluted to undetectable level. Even though ^{90}Sr in marine fishes are not detected, citizens raised questions about the safety of the fishes because of releasing the high concentrations of ^{90}Sr from the FDNPP. Therefore, in this study, we investigated the concentrations of ^{90}Sr in marine fishes collected from the coast of east Japan. The concentration of ^{90}Sr in various fishes caught around the coast of east Japan was the same as that before the accident. Although the concentration of ^{90}Sr in *Sebastes cheni* caught near the FDNPP in Dec. 2011 was 1.2 Bq/kg w.w. that of in *S. cheni* caught off Fukushima prefecture in Jun 2012 was undetectable. Therefore we suggest that the contamination of ^{90}Sr in marine fishes may be localized to a small region near the FDNPP harbor. All the results showed the concentration of ^{90}Sr in marine fishes was notably lower than that of ^{137}Cs.

1A06 Development for fish bone ash certified reference material for ^{90}Sr analysis (1) preparation of candidate material and evaluation of homogeneity

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Following the mega earthquake, tsunami, and the accident at the Fukushima Daiichi nuclear power plant on 3.11.2011, a massive amount of radioactive nuclides were released into the environment in Japan. The emergency situation required urgent measurements of radioactive isotopes in environmental samples and food samples for human health. It was required that the certified reference material of food sample for validation of measurement of radioactive isotopes because the certified reference material is essential for accurate and precise measurement of radioactive isotopes in these samples. We have developed and supplied the certified reference material of food and environmental samples. The developed certified reference materials are brown rice grain, beef flake, soybean powder, shiitake mushroom powder, in which certified massic activities (Bq kg^{-1}) of ^{134}Cs, ^{137}Cs and ^{40}K. In this paper, we described the development of certified reference material of fish bone ash (^{90}Sr, ^{134}Cs, ^{137}Cs and ^{40}K). These new certified reference materials are useful for validation of γ ray spectrometry of ^{134}Cs, ^{137}Cs and radiochemical analysis of ^{90}Sr in fish sample.

1A07 Development for fish bone ash certified reference material for ^{90}Sr analysis (2) the results of inter-laboratory experiments

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In this presentation, we will present the results of inter-laboratory experiments for certification of massic activities of ^{90}Sr in the candidate material for fish bone ash certified reference material. The 14 laboratories in Japan and 7 overseas laboratories participated on the inter-laboratory experiments. The analytical results of ^{90}Sr in the candidate material by Japanese laboratories were consistent with each other. The certified value of ^{90}Sr in the fish bone ash certified reference material was determined by the analytical results of Japanese laboratories. The analytical results of ^{90}Sr by overseas laboratories were in good agreement with the certified value.

1A08 Investigation of selective extraction of Cs, Sr by using solvent extraction II

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Previously, we discussed the ability of selective extraction of Sr(II) ion from aqueous solution by using crown ether and Perfluoro-1,11-trioxaundecane-2,10-dioic acid (PFTOUDA) as co-extractant, and HFC as diluent. In the present work, 9 compounds, 2,2,3,3,4,4,5,5-Octafluoro-1,6-hexyldiacrylate (No. 1), 2,2,3,3,4,4,5,5-Octafluoro-1,6-hexyldimethacrylate (No. 2), Dimehyldodecafluorosuberate (No. 3), Dimehylhexa decafluorosebacate (No. 4), Perfluoro-3,6,9-trioxadecanoic acid (No. 5), Dimethylperfluoro-1,10-decanedicarboxylate (No. 6), Perfluoro-1,10-decanedicarboxylic acid (No. 7), Dodecafluorosuberic acid (No. 8) and Hexadecafluorosebacic acid (No.9) were used as co-extractants with crown ethers in expectation of cooperative effects. In the experiment (1), the solubility of these co-extractans in HFC was studied in room temperature. Thought, No.7 to No.8 compounds were difficult to dissolve in HFC media, even other compounds were dissolved over 0.02 M. In the experiment (2), the solvent extraction of strontium by Dicyclohexyl-18-crown-6 ether with dissolvable 6 co-extractans as extractant, and HFC as diluent was studied.

1A09 Analysis of time-course change of Sr-90 and Cs-137 in environment by radioactive concentration in teeth of cattle as an indicator.

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The concentrations of Sr-90 and Cs-137 of the teeth of abandoned cattle that were released into the fields in the evacuation area following the Fukushima-Daiichi Nuclear Power Plant (FNPP) accident were determined. The Sr-90 and Cs-137 concentrations varied from 25 to 235 mBq (g Ca)$^{-1}$ and 1 to 8 mBq (g Ca)$^{-1}$. They showed a positive relationship to pollution degrees of the areas where the cattle resided after the accident. The Sr-90 concentrations of each tooth was different even for the same individual, and its difference was related to the tooth development stage at the time of the accident: a higher Sr-90 concentrations was found in teeth that were in the early development stage, while a lower Sr-90 concentrations was given in teeth in the late development stage. The findings demonstrate that Sr-90 is an excellent indicator to record time course of ^{90}Sr pollution in the environment. On the other hand, there is no difference of Cs-137 concentrations of teeth between early and late development stage. The mechanism of Cs-137 incorporation in teeth differ that of Sr-90 incorporation and it is required to observe for indicator to record time course of Cs-137 pollution in the environment.

1A10 Leaching Behavior of Actinide Elements from Fuel Debris

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On March 2011, the loss of coolant accident occurred at Fukushima Daiichi NPS, because of the mega earth quake and the big Tsunami. At the reactor cores, the melted fuels were thought to have reacted with Zr claddings. As a result, a solid solution of uranium and zirconium oxides called fuel debris were expected to be formed (=$Zr_yU_{1-y}O_2$). Seawater and pure water injected for emergency core cooling. Consequently, fission products leached from the fuel debris, which generated huge amount of contaminated water. In addition, limited concentration of actinide elements are detected in the contaminated water, which may give an impact for the future waste management. Since seawater contains a lot of salts, colloids and suspended matters, the leaching behavior with seawater is expected to be different from that with pure water. In this work, therefore, simulated fuel debris consisting of UO_2-ZrO_2 solid solution doped with ^{137}Cs, ^{236}Pu, ^{237}Np and ^{241}Am tracers were synthesized by a heat treatment, and devoted for an agitated leaching test in natural seawater or distilled water for comparison. After the leaching test, leaching ratios were determined by alpha or gamma ray spectrometries. Cs was leached to seawater 30~40% but the leaching ratio of actinides were very low (0.1% or less). The effect of UO_2 and ZrO_2 ratio on the leaching ratios were also investigated.

1A11 Atmosphere Dependence of Formation Process of Oxygen Vacancies in Zinc Oxide Doped with Al and In Impurities

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Thermal behavior of Al and In impurities doped in zinc oxide (ZnO) was investigated by means of the time-differential perturbed angular correlation (TDPAC) method with the ^{111}In(→ ^{111}Cd) probe. In one of our previous TDPAC studies, we observed contrasting atmosphere dependence of the stability of aggregations of ^{111}In and Al impurities doped in 100 ppm Al-doped ZnO: (i) Al and In impurities associate with each other by their thermal diffusion in air, and (ii) the ^{111}In probe is detrapped from the Al aggregations in high-temperature vacuum, resulting in substitution at defect-free Zn sites. Detailed investigation of the thermal behavior of the impurities has revealed that the dissociation reaction is triggered by the formation of oxygen vacancies. Furthermore, the formation energy of oxygen vacancy was evaluated from the temperature variation of the rate constants, which was estimated from the annealing time dependence of TDPAC spectrum. In the session, based on the further investigation of TDPAC spectra for Al-doped ZnO obtained by isochronal annealings in Ar gas atmosphere, we discuss the atmosphere dependence of the formation energy of oxygen vacancy.

1A12 First detection of a missing spinel $ZnIn_2O_4$ formed as nanostructures in ZnO

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Zinc oxide (ZnO) is an intrinsic n-type semiconductor, and its physical properties are variable with the presence of dilute impurities. For future applications of this compound, it is thus important to investigate the way they are in ZnO matrix. In a series of this work, we applied time-differential perturbed angular correlation (TDPAC) spectroscopy with the $^{111}Cd(\leftarrow^{111}In)$ probe to a study of local fields in 0.5 at.% In-doped ZnO, and found that In ions form a unique minute structure dispersed in ZnO matrix. The perturbation pattern of the TDPAC spectra obtained for the In-doped ZnO allowed us to infer that the unique structure is of a type of spinel. For the purpose of obtaining further information on the structure, in the present study, a DFT calculation was performed for the evaluation of the hyperfine parameters obtained by the above experiments. It was found that the values of the electric field gradient and its asymmetry parameter at the Cd nucleus in the energetically optimized structure show excellent agreement with the experimental values within their uncertainties. This result indicates the first synthesis and detection of a binary spinel $ZnIn_2O_4$, whose structure and property have been discussed only in theory.

1A13 Annealing-Temperature Dependent Interaction of Vacancy-Type Defects with Impurity Hydrogen in Single Crystal ZnO Probed by Means of Positron Annihilation Lifetime Spectroscopy

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Zinc Oxide (ZnO) is well known to show electrical conductivity. Because of its transparecy, therefore, single crystal ZnO is highly expected for applications to light emitting devices, transparent electrodes and so on. In general, commercially available ZnO is an n-type semiconductor because hydrogen atoms unwantedly mixed into ZnO through crystal growth process work as a donor in ZnO. It is considered that the impurity hydrogen atoms form complexes with zinc vacancy (V_{Zn}), represented by V_{Zn} + nH, by coulombic attraction between them, which could be a cause of inactivation of V_{Zn} as an important contributor to p-type semiconductivity of ZnO. The electrical conductivity of ZnO can be controlled by the variation of the concentration of the hydrogen, i.e. the carrier density, and it is thus essential to examine interacting nature of V_{Zn} and impurity hydrogen.

In this work, we introduced hydrogen ions ($^1H^+$) into ZnO single crystal and observed the state of the hydrogen atoms as impurities by means of positron annihilation lifetime spectroscopy. In the present session, we discuss annealing-temperature dependent interaction between V_{Zn} and $^{1}H^{+}$ based on results of electrical conductivity measurements as well.

1A14 Mössbauer spectroscopic study of valence-trapping/detrapping in mixed-valence trinuclear iron pentafluorobenzoate complexes, forming two polymorphs and having crystalline solvated molecules

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We have studied valence-fluctuation behaviors in mixed-valence trinuclear iron pentafluorobenzoate complex $Fe_3O(C_6F_5CO_2)_6(C_5H_5N)_3$ by ^{57}Fe Mössbauer spectroscopy. This electrically-neutral complex was synthesized with dichloromethane as a crystal solvate molecule, $Fe_3O(C_6F_5CO_2)_6(C_5H_5N)_3 \cdot CH_2Cl_2$, which was reported to be a novel compound by us in 2012, together with the Mössbauer data. It was revealed in our recent work that the $Fe_3O(C_6F_5CO_2)_6(C_5H_5N)_3 \cdot CH_2Cl_2$ complex have two polymorphs; hexagonal and orthorhombic forms. By single crystal X-ray structure analysis, in the former three bond lengths (Fe-O) and bond angles (Fe-O-Fe) in a central Fe_3O moiety were shown to be equivalent one another, while in the latter they ware proved to be non-equivalent. Mössbauer measurements showed that the room-temperature valence-state of three iron cations in the hexagonal form is detrapped, i.e., only averaged-valence iron $Fe^{2.7+}$ being observed, and that the valence-trapped state (Fe^{3+} and Fe^{2+} observed discretely) is found in the orthorhombic form at room temperature. This finding implies that the iron-valence fluctuation should be influenced by structures of the central Fe_3O moiety.

On the other hand, we developed an experimental method of replacing CH_2Cl_2 with other organic solvated molecules (Sol) in $Fe_3O(C_6F_5CO_2)_6(C_5H_5N)_3 \cdot CH_2Cl_2$, forming $Fe_3O(C_6F_5CO_2)_6(C_5H_5N)_3 \cdot Sol$, where Sol = benzene, toluene, xylene, pyridine, and so on. In the present work, the Mössbauer spectra and powder XRD patterns were measured for $Fe_3O(C_6F_5CO_2)_6(C_5H_5N)_3 \cdot Sol$, in order to clarify furthermore such relationship between the valence-fluctuation and the Fe_3O structures.

1A15 Spatial variation of the FDNPP-derived ^{134}Cs in coastal/offshore areas of Niigata

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We collected a total of 13 marine sediment core (10 cm) samples in the coastal and offshore areas (40-520 m depths) in the Sea of Japan-side of central of Honshu Island in July 2014 during the R/V *Soyo Maru* expedition and examined lateral and vertical profiles for concentrations of ^{137}Cs (half-life: 30.2 y) and ^{134}Cs (half-life: 2.06 y) in this area. The concentration of ^{134}Cs (0-7 Bq/kg-dry) at the surface (0-1 cm depth) decreased sharply towards the offshore area and along the coastline from the vicinity of an estuary of Agano River. Near an estuary of Agano River, ^{134}Cs has reached to 10 cm depth within 3 y after the Fukushima Dai-ichi Nuclear Power Plant accident.

1A16 Chemical species of cesium in suspended particles in seawater

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Chemical species of radiocesium adsorbed on sinking particles and suspended particles in seawater and that in river water was studied. In particular, their affinity to Cs was examined before and after the removal of organic matter associated with the particles by leaching experiments, Cs speciation by EXAFS, and characterization of organic matter by scanning transmission X-ray microscope (STXM). It was found that Cs-137 newly added and adsorbed to the particles can be leached by the addition of 1 M NH_4Cl solution. However, the leached fraction decreased when Cs-137 was adsorbed on the particles after the removal of organic matter from particles by hydrogen peroxide solution. Solid-water distribution coefficient (Kd) of Cs on the particles increased and inner-sphere complex of Cs in the particles increased by the removal of organic matter from the particles, showing that Cs affinity for the organic-free particles, presumably clay minerals, is high. Original Cs-137 incorporated in the particles was not leached by 1 M NH_4Cl solution both in the absence and presence of organic matter, which shows that Cs-137 was adsorbed on the particles before its migration into seawater.

1A17 Study on transport behavior of radiocesium in the lower Abukuma River after the Fukushima Dai-ichi Nuclear Power Plant accident

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The secondary radioactive dispersion of radiocesium from the contaminated watershed to the river waters is important to understand the migration behavior of ^{134}Cs and ^{137}Cs in river systems running through the areas with widely various radiocesium deposition on ground surfaces. This study investigated ^{134}Cs and ^{137}Cs radioactivity in water samples at fixed stations (Date and Iwanuma) in the lower Abukuma River, Japan after the Fukuhsima Dai-ichi Nuclear Power Plant accident.

The radiocesium activity concentration had a decreasing trend with increasing time at normal flow condition during May 2011- February 2015 after the accident, though the highest radiocesium activity concentrations were observed after rain events. The percentage of ^{134}Cs and ^{137}Cs associated with riverine suspended solids was 29-89% with an average of 70 ± 18% at normal flow condition, but ranged from 84 to 99% and an average of 96 ± 6% at high flow condition due to rain events. These results indicate that rain events are a main factor controlling the transport flux of radiocesium associated with suspended solids in river waters from the lower Abukuma River.

1A18 Relationship between radiocesium interception potential (RIP) and various factors such as cation exchange capacity, specific surface area, organic carbon content, and mineral composition for suspended sediment in river water

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Characterization of 62 suspended sediment samples collected from rivers in Fukushima and Miyagi Prefectures were conducted including solid-water distribution coefficient (Kd), radiocesium interception potential (RIP), cation exchange capacity (CEC), specific surface area (SA), organic carbon content (OC), and mineral composition (in particular, mica (M) and quartz (Q) ratio). The correlation of RIP with K_d, CEC, SA, OC and M/Q ratio were examined by multiple regression analysis. It was found that RIP is positively correlated with CEC, SA, and M/Q, while negatively correlated with OC, which was not clearly observed by 1:1 correlation of RIP to each parameter.

1B01 Measurement of the first ionization energies of fermium (Fm, Z=100) and mendelevium (Md, Z=101)

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The first ionization energies (IPs) of heavy elements with atomic numbers Z > 100 have not been measured due to their short half-lives and low production rates. In order to determine IPs of heavy elements experimentally, we have developed a novel measurement method based on a surface ionization technique coupled to a He/CdI$_2$ gas-jet transport system in the Isotope Separator On-Line (ISOL) at the JAEA tandem accelerator facility. In this work, we have determined IP values of Fm (Z = 100) and Md (Z = 101). In a surface ionization process, an ionization efficiency of an atom depends on its IP. To obtain a relationship between IP and ionization efficiency in the present system, we measured ionization efficiencies of short-lived isotopes, ^{80}Rb, ^{157}Er, ^{162}Tm, ^{168}Lu, ^{142,143}Eu, ^{143m}Sm, ^{148}Tb, ^{153,154}Ho, ^{165}Yb, and ^{49}Cr. Ionization efficiencies of ^{249}Fm and ^{251}Md, produced in the ^{243}Am(^{11}B, 5n) and ^{243}Am(^{12}C, 4n) reactions, respectively, were also measured. Measured ionization efficiencies of ^{249}No and ^{251}Md were 1.2 %. From these ionization efficiencies of Fm and Md, their IP values were determined based on the relationship between of IPs and ionization efficiencies in the present system. In the presentation, we will report on the first experimental determination of the IP values of Fm and Md.

1B02 Adsorption Behavior of Elements in Surface Ionization: Towards Measurement of Adsorption Enthalpy of Lawrencium (Lr)

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Due to strong relativistic effects, the ground state electronic configuration of the heaviest actinide, lawrencium (Lr), is predicted to be [Rn]5f^{14}7s^{2}7p$_{1/2}$, which is different from that of the lanthanide homolog Lu [Xe]4f^{14}6s^{2}5d. According to the semi-empirical calculations based on the electronic configuration, the volatility of Lr is higher than that of Lu. Here we have investigated adsorption behavior of Lr together with various short-lived lanthanide isotopes using a surface-ionization technique that are produced in heavy-ion induced nuclear reactions at the JAEA tandem accelerator. The results suggest that the adsorption of Lr on a Ta surface is smaller than that of Lu, indicating that the volatility of Lr is higher than that of Lu as expected.

1B03 Spectroscopy of low-energy conversion electrons to investigate the deexcitation process of ^{235m}U

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The ^{235m}U nuclide has very low excitation energy (76.8 eV) and decays predominantly by the internal conversion process. The deexcitation of ^{235m}U involves the interaction between the nucleus and outer-shell electrons. Indeed, the variation of the decay constant of ^{235m}U depending on its chemical environment was reported. We are aiming to clarify the deexcitation process of ^{235m}U by measuring the decay constants and the energy spectra of the internal-conversion electrons for various chemical forms. In this work, we fabricated a spectrometer of low-energy conversion electrons of ^{235m}U. The spectrometer is retarding-field type, and electrons can be detected at high efficiency. We evaluated the performance of the spectrometer by measuring auger electrons of ^{51}Cr and ^{48}V. Then we prepared ^{239}Pu sources, and tried to measure decay constants and energy spectra with the spectrometer for ^{235m}U samples collected from the ^{239}Pu sources to Cu plates by the Collection Apparatus for Recoil Products. The decay constants were determined for different ^{235m}U samples.

1B04 Search for the vacuum ultraviolet photons emitted from ^{229m}Th samples

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Thorium-229m lies in extremely low energy level, 7.8 ± 0.5 eV, and this excitation energy corresponds to vacuum ultraviolet light. Because of such a low energy, it is expected that ^{229m}Th deexcites interacting with its outer orbital electrons, and the half-life and even the decay mode of ^{229m}Th depend on its chemical environments. Recently, photon emissions from the ^{229m}Th sample implanted into MgF_2 glass were reported and its half-life was evaluated to be about 6 h. In this study, we report the results of photon detections of ^{229m}Th samples with different chemical forms: fluoride and hydroxide. Thorium-229m was separated from its mother nuclide, ^{233}U, by anion-exchange separation. Then, ^{229m}Th was coprecipitated with samarium and the photon detection was performed in the energy range 4-10 eV. For the hydroxide and fluoride samples of ^{229m}Th grown for about 2 h, no significant photon emission was observed. In the case of the ^{229m}Th samples grown for about 13 h, the photon counts decaying for time were detected. However, these decaying photons possibly originate from the daughter nuclides, such as Bi or Pb. Indeed, photons were not observed for purified ^{229m}Th samples. In addition, we tried photon detection for ^{229m}Th implanted into MgF_2 glass.

1B05 Ion-exchange experiments of Nb and Ta in HF/HNO_3 solution for identification of fluoride species of Db

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We have so far studied the anion-exchange behavior of dubnium (Db) in 14 M HF and 0.3 M HF/0.1 M HNO_3 solutions; the adsorption probability of Db was lower than those of Nb and Ta in the former solution and was close to that of Nb in the latter. In the present study, for identification of fluoride species of Db, we further studied anion-exchange behavior of group-5 element Nb and Ta in 1.0-24 M HF/2.0 M HNO_3. In the experiment, carrier-free ^{95g}Nb and ^{179}Ta radioisotopes produced in the ^{nat}Zr(*p*, *xn*) and ^{nat}Hf(*p*, *xn*) reactions, respectively, were separated from the target materials by an ion-exchange method. Batch-wise experiments with ^{95g}Nb and ^{179}Ta were carried out in 1.0 - 24 M HF/2.0 M HNO_3 using the anion-exchange resin

CA08Y. Results showed that adsorption probability of Nb on the resin sharply increases at > 6 M HF/2.0 M HNO_3 while that of Ta gradually decreases with increasing HF concentration. This behavior suggests that Nb forms $[NbF_6]^-$ and $[NbF_7]^{2-}$ from $[NbOF_5]^{2-}$ at > 6 M HF and Ta exists as $[TaF_6]^-$ and $[TaF_7]^{2-}$ in the solution. In conclusion, it is expected that chemical species of Db could be identified with the anion-exchange chromatography under the present conditions.

1B06 Determination of the distribution coefficients in the extraction of anionic chloride complexes of rutherfordium

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To investigate chloride complexation of element 104, rutherfordium (Rf), we studied extraction behaviors of its homologous elements (Zr and Hf) and pseudo homologue (Th) in an Aliquat 336/HCl system. In addition, we developed a batch-type solid-liquid extraction apparatus for online use (AMBER) to investigate time dependences of the extraction behavior of Rf and determine the distribution coefficient (K_d) of it. In this study, we performed online solid-liquid extraction experiment of ^{261}Rf and ^{169}Hf in an Aliquat 336/HCl system. These nuclides were simultaneously produced in the bombardment of ^{248}Cm and ^{nat}Gd mixture target with ^{18}O beams delivered from an AVF cyclotron at RIKEN. The products were continuously transported to the chemistry room with a He/KCl gas-jet system. We performed solid-liquid extraction of these elements with various shaking time (10–60 s) by using AMBER. After the extraction, we measured alpha-particle for the samples by using an automated rapid α/SF detection system. We succeeded in the observation of the equilibration of the extraction reaction of Rf. The obtained K_d values of Rf were clearly different from those of Zr, Hf, and Th.

1B07 Development of a flow solvent extraction apparatus coupled to the GARIS gas-jet system for aqueous chemistry of superheavy elements

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Toward aqueous chemistry of the heaviest superheavy elements such as element 106, Sg and element 107, Bh, we have been developing a rapid solvent extraction apparatus which consists of a continuous dissolution apparatus (Membrane DeGasser: MDG), a Flow Solvent Extractor (FSE), and a flow liquid scintillation detector for α/SF-spectrometry. As a preseparator for this chemistry apparatus, we plan to use the RIKEN GAs-filled Recoil Ion Separator (GARIS). In this work, we studied the performance of MDG using ^{90m,g}Nb (m: $T_{1/2}$ = 18.8 s; g: $T_{1/2}$ = 14.6 h) and ^{178a}Ta ($T_{1/2}$ = 2.36 h) produced in the ^{nat}Zr(d,xn) and ^{nat}Hf(d,xn) reactions, respectively, at the RIKEN AVF cyclotron. It was found that the dissolution efficiency of the short-lived ^{90m}Nb is around 50% while those of the relatively long-lived ^{90g}Nb and ^{178a}Ta are as high as over 80%. FSE consists of a Teflon capillary and a phase separator with a membrane filter. We also studied solvent extraction behavior of ^{95m}Tc ($T_{1/2}$ = 61 d) and ^{183}Re ($T_{1/2}$ = 70 d) into tri-n-octylamine (TOA) in toluene from nitric acid solutions. Distribution ratios of ^{95m}Tc and ^{183}Re obtained with FSE were consistent with those by a batch method.

1B08 Dependence on F^- concentration of cationic fluoride complexation of super-heavy element rutherfordium by using TTA-reversed-phase chromatography

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We have so far developed a reversed-phase-chromatography technique with TTA as a chelating extractant to clarify properties of a cationic fluoride-complex of rutherfordium (Rf). In this study, we studied elution behavior of Rf in HF/HNO_3 solutions by this technique. ^{261}Rf was produced in the ^{248}Cm(^{18}O, 5*n*)^{261}Rf reaction at the RIKEN K70 AVF cyclotron. ^{89m,85}Zr and ^{175,169}Hf, short-lived nuclides of lighter homologs of Rf, were also produced in the ^{nat}Y(*d*, x*n*)^{89m}Zr, ^{nat}Gd(^{18}O, x*n*)^{85}Zr, ^{nat}Lu(*d*, x*n*)^{175}Hf, ^{nat}Gd(^{18}O, x*n*)^{169}Hf reactions, respectively. Reaction products were rapidly transported with a KCl/He gas-jet system to the chemistry laboratory. TTA-chromatographic behavior was investigated in HF/0.01M HNO_3 solutions using the automated rapid chemistry apparatus (ARCA) and the rapid α/SF detection system for aqueous chemistry of super-heavy elements at RIKEN.

As a result, 160 α events (8.00-8.40 MeV) from ^{261}Rf and its daughter nuclide ^{257}No were observed in 1001 cycles of the column chromatography experiment. The percent adsorption value of Rf was constant at around 60% at $[F^-] < 5.0\times10^{-4}$ M and then steeply dropped at $[F^-] = 9.0\times10^{-4}$ M, while those of Zr and Hf showed decreases at $[F^-] > 4.0 \times 10^{-5}$ M. This suggests that the cationic fluoride complexes of Rf more stably exist than those of Zr and Hf at $[F^-] > 4\times10^{-5}$ M.

1B09 Extraction behavior of Mo and W from H_2SO_4 into toluene with Aliquat336 as model experiments for seaborgium (Sg)

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We have started studying sulphate-complex formation of a transactinide element, seaborgium (Sg). In this study, we report on the extraction behavior of carrier-free radioisotopes ^{93m}Mo ($T_{1/2}$ = 6.85 h) and ^{181}W ($T_{1/2}$ = 121.8 d) which are lighter homologs of Sg, from aqueous H_2SO_4 solution with amine extractant, Aliquat336, dissolved in toluene by a batch method. These radioisotopes were produced in the ^{nat}Zr(*α*, 2*n*) and ^{181}Ta(*d* ,*xn*) reactions at the RIKEN K70 AVF cyclotron. Results of the extraction experiments showed that the distribution ratios of Mo and W increase sharply above ~ 3.0 M H_2SO_4. Based on the slope analysis separately carried out, it was indicated that anionic sulphate-complex of $[MO_2(SO_4)_2]^{2-}$ (M = Mo, W) are formed in > 5 M H_2SO_4. These results suggest that the present system is applicable to the extraction of Sg. In the symposium, we will also show the present status of the development of a new solvent extraction apparatus for future Sg experiments.

1B10 Estimation of Carbon Transfer from Soil to Plant using Stable Isotope Ratio

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The aim of this study is to estimate soil carbon-14 (^{14}C, half-life: 5730 y) transfer to crop considering deep underground TRU waste disposal because ^{14}C is an important radionuclide in the radiation dose assessment. The soil to plant transfer factor (TF) is used in the assessment and TF is calculated as a ratio of concentration in crop divided by that in soil. For the case of carbon, TF was calculated from stable carbon mass so that the value was extremely high; however, it did not reflect the real carbon cycle, i.e. plant photosynthesizes CO_2 from the air, and the effect from soil to plant was not clear. Moreover, only a limited numbers of radiotracer experiment data were available. To obtain TF in the field conditions, we measured stable carbon isotope ratio, $\delta^{13}C$, in crop and associated soil samples to identify whether we could find any correlations between soil $\delta^{13}C$ and crop $\delta^{13}C$. There was no correlation for leafy vegetables and tubers, and only a low correlation was found for rice. Thus we concluded that only a negligible amount of soil carbon would be used in plants. We did further analysis and the results on estimated carbon TFs will be presented.

1B11 Effect of ashing temperature on accurate determination of plutonium in soil samples

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Pu leaching with nitric acid is widely employed to determine global fallout Pu in solid samples, i.e. soil and sediment. At sampling ashing step to decompose organic matter, various temperatures (400 – 900°C) are used. However, the effect of temperature on the accurate Pu analysis has not been well investigated. In this study, we used two standard reference soils (IAEA-soil-6 and IAEA-375) and Pu determination was carried out after ashing at various temperatures from 375-600°C. The results showed that $^{239+240}Pu$ activity was lower than the certified value when the ashing temperature exceeded 450°C, and the $^{239+240}Pu$ activity continue to decrease as the ashing temperature raised. Approximately 40% Pu could not be leached out by concentrate HNO_3 after ashing for 4 h at 600°C. The Pu loss was attributed to the formation of refractory materials, which are insoluble in HNO_3 solution. This hypothesis was confirmed by the XRD analysis of ashed soil samples which revealed that plagioclase-like silicate materials were formed after high temperature ashing. To ensure the Pu release efficiency in HNO_3 leaching, we recommend 450°C as the ideal ashing temperature.

1B12 Radiocesium accumulation and Pu isotopes source identification in sediments from Lake Inba, Japan after the Fukushima Daiichi Nuclear Power Plant accident

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To investigate the vertical distributions of ^{137}Cs and Pu in lake sediments after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, two sediment cores were collected on 11 August 2011 (Core-1) and 30 July 2012 (Core-2) at the same position in Lake Inba. Radiocesium (^{134}Cs, ^{137}Cs) were measured by a Ge-detecting system, and Pu isotopes (^{239}Pu, ^{240}Pu) were analyzed by SF-ICP-MS after radiochemical separation. The ^{134}Cs/^{137}Cs activity ratios of upper layers in the two sediment cores were approximately 1 (decay corrected to 15 March 2011), suggesting that the FDNPP accident-derived radiocesium have contaminated the samples. For Core-1, ^{137}Cs inventory was calculated to be 32300 ± 660 Bq/m^2. For core-2, collected about one year later, the ^{137}Cs inventory did not change much with the value of 37500 ± 330 Bq/m^2. The activities of $^{239+240}$Pu ranged from 0.007 to 0.298 Bq/kg, which was comparable with that of freshwater lake and marine sediments around Japan before FDNPP accident. The atom ratios of ^{240}Pu/^{239}Pu around 0.18 indicated that global fallout was the main source for Pu contamination in lake sediments.

1B13 Attachment behavior of fission products on solution aerosol

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Various characteristics such as size distribution, chemical component and radioactivity have been analyzed for radioactive aerosols released from Fukushima Daiichi Nuclear Power Plant. Measured results for radioactive aerosols suggest that the potential transport medium for radioactive cesium was non-sea-salt sulfate. This result indicates that cesium isotopes would preferentially attach with sulfate compounds. In the present work the attachment behavior of fission products on aqueous solution aerosols of various salts, such as NaCl, NaBr, NaI, KCl and CsCl, has been observed using a generation system of solution aerosols and spontaneous fission source to elucidate a roll of chemical and physical effects in a generation mechanism of radioactive aerosols. Attachment ratios of fission products on the solution aerosols were compared among the aerosols generated by different solutions. It was found that proportional relationship exists between the attachment ratios and the total surface area. In comparison of slopes of the relationship, there found a significant difference according as a solute of solution aerosols. The difference according to the solute shown in the present work suggests the existence of chemical effect in the attachment behavior of fission products on solution aerosols.

1B14 Survey of contamination of radionuclide released from the Fukushima DNPP accident - Construction of database -

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Just after an accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP), data of radionuclides released from the FDNPP were measured for environmental samples by some researchers at individual research institute and university. However a comprehensive database including these data is not constructed now. Working group of the Japan Society of Nuclear and Radiochemical Sciences decided to construct a comprehensive database of data of the radionuclides in environmental samples. This database contains data such as sampling location, sampling method, sampling times, type of measurement system, concentrations of the radionuclides and others in the publications/reports and unpublished issues. Researchers input the data in a dedicated Excel sheet. The working group verifies and registers the data in the database, and then summarizes them in a scientific report.

As the first stage of the construction of the database, data containing the concentrations of the radionuclides in airborne particles is handled. The concentrations of the FDNPP-derived radionuclides in surface air were measured at Chiba, Saitama, Ishikawa, Aichi and Fukuoka. Temporal variation of their concentrations let us estimate atmospheric motion of radioactive plume released from the FDNPP.

1B15 Production of ^{67}Cu using the ^{70}Zn($d,\alpha n$)^{67}Cu reaction

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Since 2007, we have distributed purified radioisotopes such as ^{65}Zn, ^{85}Sr, ^{88}Y, and ^{109}Cd prepared at RIKEN Nishina Center for the purpose of contribution to society throughout industrial application of accelerator based-science. Copper-67 ($T_{1/2}$ = 61.83 h, I_{β^-} = 100%) is one of the promising radioisotopes for radiotherapy and radiodiagnosis. In this work, for the future distribution of ^{67}Cu, we investigated a novel procedure to prepare the purified ^{67}Cu using the ^{70}Zn($d,\alpha n$)^{67}Cu reaction.

Radiotracers of ^{61}Cu, ^{66}Ga, and ^{69m}Zn were produced in the ^{nat}Zn(d,X) reactions with a 24-MeV deuteron beam delivered from the RIKEN AVF cyclotron. After the irradiation, we developed the two-step chromatographic separation with Eichrom Cu resin and Dowex 1X8 anion-exchange resin. A chemical yield of Cu was 93%, and decontamination factors of Ga and Zn were > 2.6×10^4 and > 1.6×10^4, respectively. A separate irradiation was performed for an enriched ^{70}ZnO target (^{70}Zn isotopic abundance: 96.87%; target thickness: 327 mg/cm^2) in order to evaluate a production yield of ^{67}Cu and a quality of the purified ^{67}Cu. Under the present experimental condition, the yield of ^{67}Cu was 4.0 MBq μA^{-1}h^{-1}. We estimated that about 1 GBq of ^{67}Cu could be distributed after a 3-days irradiation with a 10-μA deuteron beam, followed by 3 days for chemical separation and shipment.

1B16 Production and Purification of Medical Radioisotope Y-90 with accelerator neutrons by deuterons

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A new system has been proposed for the generation of radioisotopes with accelerator neutrons by deuterons, especially the production of ^{90}Y ($T_{1/2}$ = 64 h), which has been used for the nuclear medicine. Enriched ^{90}Zr nitrate samples were irradiated with neutrons, which were obtained by the ^{nat}C(d,n) using 40 MeV deuterons provided from the TIARA cyclotron. ^{90}Y was successfully produced via the (n, p) reaction, and separated from the Zr nitrate target by three columns method with a hydrochloric acid. The chemical procedure was done within 6 hours, and the chemical yields was about 85%. The separation factor between Y and Zr was obtained to be <2x10^{-5} by using a γ-ray spectrometry. After the separation 90% of the target material was recovered from the column with 4 M HNO_3 solution for the next irradiation. After the purification, a labelling of DTPA with ^{90}Y had been performed. In this result, the yield was about 90% under >2.5 μM of DTPA, although the yields were decreased at the DTPA concentration below 2.5 μM.

1B17 Studies on PET imaging of cancer using Cu-64 ions

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Copper-64 (^{64}Cu) is an attractive radionuclide for positron emission tomography (PET) imaging due to its half-life (12.7 h) and decay characteristics. We proposed a new approach to the PET imaging of cancer using ^{64}Cu ions without any labeling process. In this study the usefulness of ^{64}Cu ions as a PET imaging agent was evaluated.

No-carrier-added ^{64}Cu was produced by the nuclear reaction of ^{64}Ni(p,n)^{64}Cu at high specific activity, using the AVF cyclotron of Takasaki Ion Accelerators for Advanced Radiation Application facility at Japan Atomic Energy Agency.

The twelve kinds of cancer cells routinely cultured were each incubated with ^{64}Cu ions in the medium at 37ºC. After incubation the cells were thoroughly washed and their radioactivity was measured. The specific cellular uptake of ^{64}Cu ions was observed in the several kinds of cancer.

The cancer-bearing mice were prepared by subcutaneous injection of the cancer cells. The mice were subjected to a small-animal PET imaging at different time points after injection of ^{64}Cu ions via the tail vein. The biodistribution study was further performed. It was demonstrated that the cancer could be successfully visualized on the PET images due to the quite high uptake of ^{64}Cu in the cancer tissue.

1B18 DNA strand break caused by charge and vibrational energy transfer between nucleotides: Reaction dynamics approach

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DNA is an important substance that carries genetic information of living organisms. When DNA is exposed to intense radiation, the DNA strands break. This may cause a wrong transcription of its base sequence and eventually harmful influence on a human body. The concern on radiation-induced DNA strand break is growing rapidly but its mechanism remains unclear at a molecular level, leading to difficulties in scientific discussions on radiation damage.

Various model DNAs which have a small number of base pairs and two linkers bridging the ends of the short strands have been synthesized as prototypes of DNAs. In this work, we performed chemical reaction dynamics simulations for short-strand DNAs to elucidate the mechanisms of strand breaks. For example, we optimized the structure of a model DNA composed of 4 adenine-thymine base pairs and gave initially a thermal energy of 0.4 eV/atom excluding linkers. We found that single-strand breaks, which mainly occur on the thymine side, consisted of two bond-breaking processes:

(i) Hydrogen transfer from a sugar and to an adjacent phosphate followed by cleavage between the sugar and its adjacent thymine
(ii) Cleavage between the sugar and the phosphate induced by energy and charge transfers from other nucleotides

2A01 Production of neutron-deficient rutherfordium isotopes in the ^{208}Pb + ^{48,50}Ti reactions

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In order to clarify a nuclear fission

mechanism, our group is planning a systematic study of fission characteristics of Rf that has relatively a large number of spontaneously fissioning isotopes. In this study, we measured decay events of especially neutron-deficient Rf isotopes under low background.

Experiments were performed at the RIKEN linear accelerator (RILAC) facility using a gas-filled recoil ion separator, GARIS. Rf isotopes were produced in the reaction of ^{208}Pb + ^{48,50}Ti. ^{208}Pb targets of approximately 300 μg cm^{-2} in thickness were prepared by vacuum evaporation on a 60 μg cm^{-2} carbon foil. The beam energies of ^{48}Ti and ^{50}Ti were 224.7–239.2 MeV and 203.5–270.0 MeV, respectively. The average intensities of ^{48}Ti and ^{50}Ti were 350 pnA and 570 pnA, respectively. The sixteen targets were put on a wheel of 30 cm in diameter and the wheel was rotated at 2000 rpm. In this experiments, we adopted a target ID system to simultaneously observe reactions at four beam energies using Al degraders (0, 0.8, 2.0, and 3.0 μm) which were installed on the beam upstream of the targets. Rf nuclides produced in the ^{208}Pb(^{48}Ti, xn) and ^{208}Pb(^{50}Ti, xn) reactions were separated from the beam particles and nuclear reaction byproducts using GARIS, and were measured with a focal plane detector box composed of a position sensitive detector (PSD) and four silicon detectors (SSD). A preliminary analysis shows that some events of ^{253}Rf and ^{254}Rf were observed in the reaction of ^{208}Pb + ^{48}Ti.

2A02 Alpha-decay spectroscopy of ^{261}Rf

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Alpha-gamma coincidence and high-resolution alpha fine-structure measurements were performed for the alpha decay of ^{261}Rf. The nucleus ^{261}Rf was produced with the ^{248}Cm(^{18}O,5n) reaction at the JAEA Tandem accelerator facility. Reaction products were transported with a gas-jet transport technique to a rotating-wheel alpha-gamma detection system. Two gamma lines associated with the alpha decay of ^{261}Rf were clearly observed, and more precise alpha-energy spectrum than before were obtained. It was found that the literature value (8.28 MeV) of the alpha-transition energy of ^{261}Rf was incorrect, and that the alpha-decay scheme of ^{261}Rf should contain several low-energy cascade gamma transitions. On the basis of these results and literature data of ^{261m}Rf , we will discuss nuclear structure of superheavy nuclei around this region as well as the level structure of ^{261}Rf and its daughter nucleus ^{257}No.

2A03 Study on hot fusion reaction of ^{248}Cm+^{48}Ca→^{296}Lv* using GARIS

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In 2013, a gas-filled recoil ion separator was employed to study the production of Lv (Z=116) isotopes in the hot fusion reaction of ^{248}Cm+^{48}Ca at the beam energy E_{labo} = 261.6 MeV. As a result, five correlated decay chains were observed. On the basis of the assignments of the precedent studies by both Dubna-Livermore collaboration and GSI SHIP collaboration, one of the events was attributed to the decay of ^{293}Lv (3n), and three of them were assigned to the decays of ^{292}Lv (4n).

With the aim of clear identification of these nuclides, we started to measure the excitation function. The ^{248}Cm target was irradiated with a 258 MeV ^{48}Ca^{11+} beam from RIKEN heavy-ion linear accelerator. Total beam dose was 5.0×10^{18} during a net irradiation time of 14.2 days. As a result, two correlated decay chains were observed. One of the events is attributed to the decay of ^{293}Lv. The other is not able to assign clearly, although it is decay chain originating from the reaction of $^{248}\mathrm{Cm}+^{48}\mathrm{Ca}\rightarrow^{296}\mathrm{Lv}^*$.

We will report about the decay properties of observed chains and the excitation function including performance of background level and transmission of GARIS for hot fusion reaction.

2A04 Measurement of (g,p)/(g,n) reaction yield ratios and comparison of the results with calculated values

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The (g,p) reactions play an important role in the production of carrier-free radioisotopes from targets irradiated with bremsstrahlung. However, there is not enough experimental data on the cross sections of the (g,p) reactions to estimate the amount of radioactivity produced in the bremsstrahlung irradiation. In this study, we have compared the experimental and calculated reaction yield ratios of (g,n) to (g,p) reactions which occur from a same nuclide to make sure that the cross section prediction code is used for the estimation of the amount of the photonuclear reaction products. The bremsstrahlung irradiations to the metal or oxide targets of $_{28}$Ni, $_{38}$Sr, $_{40}$Zr, $_{44}$Ru, $_{50}$Sn, $_{56}$Ba, $_{58}$Ce, $_{60}$Nd, $_{64}$Gd, $_{70}$Yb, $_{74}$W, $_{78}$Pt were carried out using the linear accelerator of Tohoku University. The accelerator was operated at electron energies of 30 and 50 MeV with a mean current of around 0.03 mA. After the irradiation, those targets were subjected to gamma-spectroscopy to determined (g,n) and (g,p) reaction yield. We observed a wide range of (g,n)/(g,p) reaction yield ratios from 0.45 for ^{58}Ni to 127 for ^{176}Yb at 50 MeV irradiation. The comparison between the experimental and calculated values is still in the process of analysis.

2A05 Half-life of Technetium-98

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The half-life of ^{98}Tc was re-measured using the isotope dilution method for determining the number of atoms, and γ-ray spectrometry for determining the absolute disintegration rate. The proposal half-life is $6.2\pm0.6\times10^6$ y. The branching ration of EC to β^- decay for ^{98}Tc estimated to be less than 0.3% from the γ-ray spectrometry.

In this work, two kind of samples were prepared. First sample is a ^{99}Tc source (37MBq) purchased from Amersham Co. (TCS1). It was prepared from fission products and contained ^{98}Tc. And the other sample was produced by ^{97}Mo(d, n)^{98}Tc reactions. A enriched ^{97}Mo(92.9%) self-supporting target was irradiated by the 14Mev d beam for 12 hours in the cyclotron.

2A06 Elemental analysis of clay minerals sample using Prompt Gamma ray Activation Analysis method.

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(JAEA, NIST[1])

Research on desorption mechanism of cesium ions in the clay mineral has been progress on decontamination of radioactive cesium in

Fukushima. In this investigation, trace elements in clay minerals samples were determined using prompt gamma ray activation analysis method which is very sensitive multi-elemental simultaneous determination method. The experiments carried out at the research reactor in the National Institute of Standards and Technology (NIST). 25 clay mineral samples including 6 standard samples were analyzed, and 16 elements in the samples were determined. The correlation of these elements will be discussed in the conference.

2A07 Feasibility of time-of-flight prompt gamma-ray analysis of a radionuclide sample

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The Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI), which is located at the beamline No. 04 at the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC), has been designed and developed for prompt gamma-ray analysis (PGA), nuclear cross-section data measurement, and nuclear astrophysics.

We have developed a novel technique which combines PGA with time-of -flight technique (TOF-PGA), and attempted to apply TOF-PGA to the specific detection of radioactive isotopes in simulated samples of platinum group metals in four-group partitioning process. The Ge detector array, a combination of two cluster-Ge detectors, seven large coaxial-Ge detectors and BGO anti-Compton shields surrounding cluster-Ge and coaxial Ge detectors, is installed at the flight path length of 21.5 m in ANNRI, and is used for measurement of the simulated samples. Pure metal samples of platinum group provided clear peaks on the PGA and TOF spectra. On the other hand, a large number of peaks caused interference with other peaks in simulated samples. In this study, we will provide more details of analysis and experimental results.

This work was supported in part by Grants-in-Aid for Scientific Research (25246038).

2B01 Possibility of radiocarbon dating to bronze implement: Application to the metal fittings installed in the rafter of main shrine of Izumo-taisha

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Verdigris is rust generated in bronze implement. It is produced from Cu contain in bronze and CO_2 in the atmosphere. Once verdigris is formed, it has the property which prevents formation of a new verdigris more. If extraction and radiocarbon dating of the carbon contained in verdigris become possible, the age in which the rust was formed is obtained, and also estimation of the age for which bronze ware was used may be realized. In this study, therefore, we searched for the condition of carbon extraction from verdigris. Verdigris is decomposed by heating in an evacuated glass tube. As the result, it became clear by heating at 1 h and 250℃ or more that CO_2 was extracted with 90% or more of yield. Next, the verdigris samples were extracted from the metal fittings installed in the rafter of main shrine of Izumo-taisya and the radiocarbon dating of them was performed. Between the calibrated radiocarbon age and age when they were made, there was a time lag of only 4 years. This result shows that the time lag can be neglected and that verdigris is a suitable sample for radiocarbon dating.

2B02 Stream-water organic carbon export

from a mountainous forested catchment

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(Japan Atomic Energy Agency)

Stream-water organic carbon export is an important pathway for carbon (C) loss from mountainous forest ecosystems and thus understanding the seasonal changes, mechanisms, and sources of C export are necessary. We continuously monitored dissolved organic carbon (DOC) and particulate organic carbon (POC) in a small river in a mountainous forest in Ibaraki, Japan, using a monitoring system we developed previously.

POC samples were collected monthly with cartridge filters installed in the system, which were separated by particle size and then analyzed for C and nitrogen isotopes. DOC concentrations were determined based on UV absorbance measured continuously for passing water in the system and a close correlation between DOC concentration and the UV absorbance.

In general, DOC concentration increased with increasing water flow in the river. However, the degree of the increase in the concentration varied depending on the amount of rainfall and the preceding rainfall pattern. The plot of DOC concentration vs. water discharge showed two typical shapes: (1) the DOC concentration increased with the water discharge, but plateaued during high water discharge; and (2) the DOC concentration showed a sharp, linear response to the water discharge and reached a higher concentration than in (1). These different responses were probably caused by the differences in rainwater routes to the river.

2B03 Development of uranium age-dating using ^{234}Th/^{230}Th ratios

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The uranium age-dating is a nuclear forensics analysis techniques, by measuring the ^{230}Th / ^{234}U isotope ratio in uranium sample, to estimate the elapsed time from being separated and purified. We have determined ^{230}Th and ^{234}U using isotope dilution mass spectrometry method, it has conducted for uranium age-dating. Act to perform the isotope analysis after the addition a known quantity of the isotope (spike) to the sample, after spiking, analytical sample is assumed to be in the state of isotopic equilibrium. However, the samples which have complex speciation of uranium and thorium are analyzed, may not sufficiently reach to the state of isotopic equilibrium has been suggested from the experimental results. In this study, without the addition of a spike, from the measurement results of the uranium isotope ratios and thorium isotope ratio in the sample, it was investigated a method for calculating the ^{230}Th / ^{234}U isotope ratio.

The state of radioactive equilibrium between ^{238}U and ^{234}Th in the sample was utilized in the ^{234}Th / ^{230}Th method. The ^{230}Th / ^{234}U ratio was calculated using measured ^{234}Th / ^{230}Th isotope ratio, ^{234}U / ^{238}U isotope ratio and ^{234}Th / ^{238}U ratio in the radioactive equilibrium.

2B04 Search for supernova signature of ^{60}Fe

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Iron-60 with a half-life of 2.60 My is produced in the explosive environment with high neutron density such as core collapse supernovae. The nuclide is hardly produced by nuclear reactions with cosmic rays in atmosphere. The Fe-60 atoms were present in the early solar system but now extinct because of its short life time. Hence, presence of the live Fe-60 indicates signature from supernovae which occurred at vicinity of the solar system. Knie (2004) detected the ^{60}Fe in a manganese crust collected at the eastern Pacific. We attempted to detect the ^{60}Fe from manganese crusts, nodules, and deep-sea sediment collected from different location to verify the previous work.

Ages of the crusts were determined from the ^{10}Be/Be ratio. In measurement of the ^{60}Fe with an AMS, we have achieved detection limit of 4×10^{-17} for the ^{60}Fe/Fe ratio. We found a ^{60}Fe peak with the ratio of $(2–3)\times10^{-15}$ from both crust and sediment samples in layers with ages of 1.7–3.2 Ma. Another peak was observed in layers with age of 6.5–8.7 Ma from one crust sample. The enhancement in ^{60}Fe is thus observed in 1.7–3.2 Ma from all samples collected from different location.

2B05 Ligand strategy for the separation of minor-actinides and its theoretical evaluation

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We have investigated the bonding state and stability of Eu / Am complexes in order to achieve the Partitioning and Transmutation strategy of high-level radioactive waste. Our previous work by means of DFT calculation revealed that the contribution of f-electron to the bonding property depending on donor atoms determines the Eu / Am selectivity. We aim to introduce the effective donor group into a TPEN ligand to give a higher selectivity of Am. The present study evaluates the performance of a carbonyl donor to Eu / Am separation with a diglycolamide (DGA) type ligand. The calculation model, $[M(DGA)(H_2O)_6]^{3+}$, was prepared by the modification of X-ray coordinates of $[M(H_2O)_9]^{3+}$. We assume that the relative stability between Am and Eu complexes in aqueous solution corresponds to the Eu / Am selectivity. The carbonyl oxygen donor as an ester selects Eu(III) ion, whereas, the carbonyl oxygen donor as a ketone or an amide selects Am(III) ion. These results imply that the modification of carbonyl group enables us to control the Eu / Am selectivity.

2B06 Structural transformation of cyclouranyl(VI) complex by addition and elimination of proton

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In the present study, the μ_3-O and μ_3-OH bridged trinuclear uranyl(VI) complexes, $(Et_3NH)_2[(UO_2)_3O(t\text{-}Busap)_3]$ (**1**) and $(Et_3NH)[(UO_2)_3OH(t\text{-}Busap)_3]$ (**2**) were synthesized by the reactions of $UO_2(NO_3)_2\cdot6H_2O$, 1 equiv. of t-BusapH$_2$ and triethylamine (TEA) in THF. The complex **1** was obtained by the addition of 3 equiv. of TEA, and **2** was afforded by 2 equiv. of TEA. The structures of **1** and **2** were determined by single crystal X-ray diffraction analysis. The U···U distance is av. 3.83 Å for **1** and av. 3.97 Å for **2**, respectively. The U-μ_3-O distance in **1** (av. 2.23 Å) is much shorter than that in **2** (av. 2.43 Å). The U-μ_3-O-U angle in **1** is 117~119°, suggesting that the μ_3-O is sp^2 type. On the other hand, the U-μ_3-O-U angle in **2** is 110°, which indicates that μ_3-O in **2** is sp^3 type. With the help of DFT calculations, we characterized that **1** is μ_3-O bridged and **2** is μ_3-OH bridged complexes, respectively. The transformation reactions between **1** and **2** were investigated.

2B07 Electrode Reaction of Np in a weakly acidic solution

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Electrolytic reactions of Neptunium ions in a weakly acid solution were investigated by using gold electrode. It was reported previously that electrolytic reduction of Neptunium (V) ion based on mediator reaction with Np(III)/Np(IV) couple was observed in the acidic solution when bulk electrolysis was carried out at the potential around -0.2 V versus Ag/AgCl reference electrode. The reduction of Np(V), however, was not observed at the pH higher than 2. This is attributable to low reaction rate of electron exchange reaction between Np(V) and Np(III). Electrode reaction of Np(V) at more negative potential was investigated by cyclic voltammetry. The current peak due to reduction of Np(V) was observed at ca. -0.75 V, and oxidation current was appeared at +0.5 V. The reactions was determined to be reduction of Np(V) and oxidation of deposited species of reduction product of Np on the electrode. It was found that stripping current due to oxidation of deposit was saturated in spite of longer preelectrolysis time. This phenomenon is different from uranium.

3A01 Optimisation of the production parameters of ^{211}At via the ^{209}Bi(^{7}Li, 5n)^{211}Rn reaction

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Radon-211 ($T_{1/2}$ = 14.7 h), which is a parent of an alpha-emitter ^{211}At ($T_{1/2}$ = 7.2h), is a key isotope for R&D of targeted alpha therapy using ^{211}At. In order to make a ^{211}Rn with an appropriate irradiation condition, herein we determined the cross sections of ^{209}Bi(^{7}Li,5n)^{211}Rn, ^{209}Bi(^{7}Li,6n)^{210}Rn and ^{209}Bi(^{7}Li,p5n)^{210}At reactions. Irradiation was carried out with ^{7}Li^{3+} beams of 60 MeV from the JAEA tandem accelerator at Tokai. Bismuth targets, aluminum backing and cover sheets were placed at a water-cooled Faraday cup. Beam current was controlled to be approximately 150-200 nA during the irradiation of 2-3 hours. The beam current was collected by the Faraday cup and was monitored on-line by using a current integrator module. The excitation function of ^{209}Bi(^{7}Li,5n)^{211}Rn reaction is in good agreement with the previously reported values by Meyer and Lambrecht, and it exhibits the maximum cross section of 733.8 ± 6.7 mb at 52.4 ± 0.3 MeV. On the other hand, the excitation function of ^{209}Bi(^{7}Li,p5n)^{210}At reaction, which was deduced as an independent yield, was lower than the previously reported one, which was measured as a cumulative yield by Meyer and Lambrecht.

3A02 Production of ^{211}At for alpha radiotherapy via ^{209}Bi(^{7}Li, 5n)^{211}Rn

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A ^{211}Rn/^{211}At generator has been proposed to supply a radiotracer ^{211}At with a half-life ($T_{1/2}$ = 7.2 h) for medical applications. To establish a technology of the ^{211}Rn/^{211}At generator, ^{211}Rn ($T_{1/2}$ = 14.7 h) has to be produced through the ^{209}Bi(^{7}Li, 5n)^{211}Rn reaction without melting bismuth metal targets due to heat produced by the irradiation of 60 MeV ^{7}Li ion beams. In this work, we have studied the effects of a He gas cooling system of an irradiation chamber constructed for the production of ^{211}Rn.

Irradiations were carried out at the JAEA tandem accelerator using the irradiation chamber equipped with the He gas cooling system. The results show that the He gas effectively cools the targets and the cooling efficiency is nearly independent of the pressure (> 1 kPa) and the flow rate (~20 L/min) of the He gas. Increase in temperature of the bismuth targets was slightly large compared with that of the aluminum target owing to the difference of their thermal conductivity.

3A03 Developments of quantitative imaging system for alpha-emitting radioisotope ^{211}At

SEGAWA, M.[1], NISHINAKA, I. [1], INOUE, T. [1], HASHIMOTO, K. [1], KURETA, M. [1]
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Radio nuclide therapy using α-particle emitted from ^{211}At is of current interest because a-particles have higher LET. Recently, the establishment of effective technique to generate ^{211}At is required. Here, quantitative imaging techniques to analyze the ^{211}At and other radioactive materials generated at the same time will be great help for the establishment of effective technique. Thus, we has developed quantitative imaging system adjusted for relatively low activity a-particles of ^{211}At which is generated in the research phase. The present system consist of α-particle scintillator, industrial camera, macro-lens and control computer. ^{211}At was generated by ^{209}Bi (^{7}Li, 5n) ^{211}Rn ➔ ^{211}At reaction at Japan Atomic Energy Agency. The consecutive images were obtained with use of ^{211}At as sample. The brightness on ^{211}At images decreased with increase in the measurement time. The deduced half-life of ^{211}At from obtained images agreed with the known value (7.22 h) within 5% error. We confirmed that the present system could visualize the consecutive images for the decreasing ^{211}At sample with great accuracy.

3A04 Effects of indirect action by Gamma-rays in Tardigrades

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The tardigrades belong to a group which have a close relationship to an arthropoda and are aquatic animal with a length of 1mm or less. Tardigrades are with remarkable abilities to withstand harsh physical conditions such as dehydration or exposure to harmful highly energetic radiation. An animal in such a condition is called a tun. But aquatic tardigrades have hight radiation tolerance. We investigated the effects of indirect action by Gamma-rays in Tardigrades(*Milnesium tardigradum*). We found that aquatic tardigrades withstand 6500Gy Gamma-rays and be affected by indirect action.We found temperature effect and dose effect. H_2O_2 concentration is affected by survival rate from 60mM to 80mM.

We will investigate effct of indirect action in Tardigrades.

3B01 Attempt of Total Synthesis for ^{14}C labeled C_{60}

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(Tokyo Metropolitan Univ.)

Recently, C_{60} and their derivatives are noted as an active ingredient owing to high radical scavenging ability and anti-HIV activity. Considering about the application to the pharmaceuticals and cosmetics, it is necessary to study the pharmacokinetics of C_{60}. A radioactive-tracer method is one of the most effective method for such research. For the synthesis of ^{14}C labeled C_{60} (C_{60}[^{14}C]), the arc discharge method using carbon electrodes adsorbing ^{14}C labeled compounds is employed. However, this method has many problem such as the inconsistency in the specific radioactivity of the produced C_{60}[^{14}C] and resulting large amount of radioactive soot as a byproduct. In the recent years, the synthetic approach to the production of C_{60} has been studied by many organic chemists. If it is possible to apply these approaches to the production of $C_{60}[^{14}C]$ some problems mentioned above would be solved. In this session, we will report the attempt of total synthesis $C_{60}[^{14}C]$.

3B02 Synthesis and Characterization of Promethium Endohedral Metallofullerenes

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Endohedral metallofullerenes (EMFs), which are fullerenes including metal atoms, have attracted interest because of their unique physical and chemical properties. It is known that various metal atoms, especially for lanthanides without Promethium (Pm), are included in the fullerene cage. Pm is known as an artificial radio element whose atomic number is smaller than 82. The chemical properties of Pm are considered to be almost same as neighboring lanthanides of Nd and Sm. However, it has been reported that the oxidation state of Sm and Nd in the fullerene cage are +2 and +3, respectively, although the most stable oxidation state is +3 in aqueous solution. Considering about the chemical properties of Pm, it is very important to investigate the valence state of Pm in fullerene cage.

In order to study the properties of $Pm@C_{2n}$, we produced Pm by photo nuclear reaction and synthesized $Pm@C_{2n}$ with DC arc discharge method. As a result of HPLC analysis of $Pm@C_{2n}$, it was found that oxidation state of Pm in the fullerene is +3 from the similarity of HPLC behavior to other metallofullerenes encapsulating trivalent lanthanide metal atom.

3B03 Improvement in production yield of radioactive strontium endohedral fullerene using nuclear recoil

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Considering about the radio pharmaceutical application of metallofullerenes, the production methods using nuclear recoil phenomena is very useful because specific radioactivity of products is higher than that obtained by other methods. Due to these advantages, this production method has been attracted and examined using various nuclear reactions so far. However, reported production yields of radioactive metallofullerenes were not enough because of the radiation damages of a targeted fullerene molecule caused by incident charged particles. One of the promising method to improve the production yield of radioactive metallofullerenes is ion implantation using the nuclear recoil induced by nuclear reaction with fast neutrons. In this study, we will report the synthesis of ^{85}Sr endohedral fullerene using the nuclear recoil of ^{86}Sr (n, 2n) ^{85}Sr reaction with 14 MeV fast neutron produced by DT reaction.

As a result, the radioactivity of ^{85}Sr observed in the aniline solutions derived from ^{85}Sr endohedral fullerene was estimated to be about 6.8% at the best for those before extraction. Generally, production yield of metallofullerenes by the standard arc discharge method is about 0.1% or less. Therefore, observed production yield of ^{85}Sr endohedral fullerene in this study is found to be extremely efficient compared to the standard methods.

3B04 Liquid scintillation counting method with little liquid organic waste

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Low energy beta-ray emitters are generally measured by a liquid scintillation counter (LSC) because of its high counting efficiency. However, the LSC method has a demerit for generating liquid organic wastes after measurement. To avoid generation of liquid organic wastes, plastic scintillators (PSs) has been used for measurement of tritium compounds with a conventional LSC. PSs are a sheet type for nonvolatile compound and pellets type for volatile compound like HTO. 0.5-mm-thick PS-sheet of BC-400 (Saint-Gobain) was cut in 13 mm width and 50 mm height. A PS-pellet of EJ-200 (G-tech Co.) is approx. 3 mm in a diameter and 3 mm in a length. Both types of PS were used to check stable counting efficiency with elapsed time, and linearity between activities and count rate. For example, the counting efficiency of ^{3}H-methionine with the PS-sheets was 43.6 ± 0.3%, which was same as liquid scintillator use. Also, the counting efficiency of HTO with the PS-pellets was 48.1 ± 0.9%, which was higher than that of liquid scintillator. Additionally, both PS could be reused by rinsing after measurement. So, there were little wastes. However, these methods had a limit for applying a large sample volume.

3B05 Nuclear Forensics and Radiochemistry

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Nuclear forensics is the analysis of intercepted illicit nuclear or radioactive material and any associated material to provide evidence for nuclear attribution by determining origin, history, transit routes and purpose involving such material. Because the nuclear forensics methodologies provide hints of the origin of the nuclear materials used in illegal dealings or nuclear terrorism, it contributes to identify and indict offenders, hence to enhance deterrent effect against such terrorism. In the 59th Symposium on Radiochemistry, the results of research and development of fundamental nuclear forensics technologies (analysis of isotopic composition, impurity analysis, age dating of nuclear material, particle shape analysis by electron microscope, and nuclear forensics library) performed in Japan Atomic Energy Agency are reported. Nuclear forensics laboratory is one of the components for nuclear forensics regime, where the facilities for analysis and storage of seized samples are included. Quality assurance system for the laboratory is needed in order to certify the reliability of the analyzed data as evidence. Japan's capability on nuclear forensics and the role of radiochemistry are also discussed in the Symposium from the points of view of international nuclear security, scientific knowledge necessary to infer the nuclear attribution by interpretation of the characteristics of nuclear and radioactive materials, and cultivation of nuclear forensics specialists.

P01 Development of correction methods for time-of-flight prompt gamma-ray analysis at ANNRI

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The Accurate Neutron-Nucleus Reaction measurement Instrument has been available for time-of-flight (TOF) prompt gamma-ray analysis. However, the specific relationship between the number of detected events and the sample weight is not entirely clear. In this study, a set of standard Fe and Ag samples were measured to investigate the factors which affect the gamma counts and TOF counts. It is found that live time of acquisition (τ), data acquisition efficiency in live time (δ), neutron attenuation (NA) and gamma-ray attenuation (GA) are the main contributing factors. An empirical formula is proposed for the calibration of δ. Simulation models are built for the evaluations of NA and GA by the PHITS program. The values of corrected gamma and TOF counts are simply proportional to the sample weight. An accuracy of typically 5% has been achieved for the corrected result of a standard sample. It is sufficient for quantitative analysis.

P02 A study on element concentration and acid neutralization capacity of soil at urban areas

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In this study, in order to tell the difference of acid neutralization capacity (ANCc) in urban areas and the proportion of ANCc in the total element concentration in the soil, we took soil samples from urban areas such as Beijing, Shanghai and Tokyo. To compare with urban areas, we also took soil from natural areas such as Yakushima and Mount Takao. Instrumental neutron activation analysis (INAA) was used to measure the total Ca, Mg, Na and K in soil. We used inductively coupled plasma atomic emission spectroscopy (ICP-AES) to measure the ANCc of soil.

Although plenty of Ca, Mg, Na and K are found in soil, over 90% exchangeable cations came from Ca and Mg in all samples while Na and K made little contribution to the ANCc. The result shows that Beijing had the biggest ANCc among 3 cities because a lot of loess moved to Beijing through dust storm in North China. On the contrary, soil in Yakushima was made of granite so that it was lack of both ANCc and the total element concentration. According to the organic compound content in soil, we can see 2 different ways of supplement between urban areas and natural areas. In urban areas (especially Beijing), ANCc revive through inorganic salts primarily. But in natural areas, ANCc revives through the leaves of deciduous trees.

P03 Cross section measurement of ^{142}Nd(p,γ)^{143}Pm and ^{143}Nd(p,n)^{143}Pm reactions at astrophysically relevant energies: Study for understanding isotopic abundance of p-nuclei ^{144}Sm

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The majority of heavy elements which are present in the solar system now were synthesized by neutron capture reaction. However, there is a group of neutron deficient nuclei, the p-nuclei, which cannot be created in such reactions. We have attempted to clarify abundance of the p-nuclei with a mass number of 130–150 by proton capture reactions. In the present work, we determined cross sections of ^{142}Nd(p,γ) and ^{143}Nd(p,n) reactions which are pathway for the nucleosynthesis of p-nuclei, ^{144}Sm. In addition, abundance of the ^{144}Sm in stellar environment was estimated with a network calculation employing evaluated cross sections.

Targets of ^{142}Nd and ^{143}Nd were individually irradiated with low-energy proton beam. We obtained experimental cross sections of ^{142}Nd(p, γ) and ^{143}Nd(p, n) in energies between 4.1 and 7.6 MeV. The experimental cross sections of (p, γ) reaction showed 3 times greater values at lower energies than theoretical prediction using TALYS code. On the other hand, the (p, n) cross sections correspond well with the theoretical prediction.

Abundance of ^{144}Sm created by the proton capture reactions on pre-existing s-seed nuclei of neodymium in explosive environment was estimated with a network calculation. If there is proton with a density of 1000 g/cm^3 at a temperature of 1.5×10^9 K for example, evaluated cross sections provide duration of 3 seconds for the nucleosynthesis.

P04 Utilization of $BiCl_3$ as standard for gas phase chemistry of superheavy elements

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In the study of super heavy elements, a technique of an isothermal gas chromatography has often been used to determine the value of an adsorption enthalpy of volatile compounds. In many cases, the adsorption enthalpy is derived from the Monte Carlo simulation which simulates behaviors of molecules in a column under certain conditions to reproduce experimental results. However, the enthalpy has more or less uncertainty, because it is difficult to estimate exact apparatus constants, experimental conditions, etc. In this study, we consider whether volatile $BiCl_3$ can be utilized as a standard compound in an isothermal gas chromatography, because ^{212}Bi, descendant in thorium series, is allowed to use for a tracer. It was found that $BiCl_3$ is volatile and pass through an isothermal column at a suitable temperature. We will report the results of macro and tracer-scale experiments and discuss more detailed chromatographic behavior of $BiCl_3$.

P05 Solvent extraction behavior of Zr and Hf with HDEHP for aqueous chemistry of Rf

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Studies on chemical properties of rutherfordium (Rf) have been carried out mainly using inorganic ligands such as halide ions, while few experiments with organic ligands have been reported. In this study, we investigated solvent extraction behavior of zirconium (Zr) and hafnium (Hf) as lighter homologs of Rf using di(2-ethylhexyl)phosphoric acid (HDEHP) as a chelate extractant. Radiotracers of ^{88}Zr ($T_{1/2}$ = 83.4 d) and ^{175}Hf ($T_{1/2}$ = 70 d) were produced by irradiating ^{89}Y and ^{nat}Lu targets with proton or deuteron beams using the AVF cyclotrons at Tohoku University and RIKEN. Aqueous solution of 1 M $HClO_4$ containing the radiotracers was mixed with an equal volume of HDEHP in toluene, and then the mixture was shaken for 2 hours. After centrifugation, the aliquot of each phase was subjected to γ-ray spectrometry using Ge detector. In general, the distribution ratio (D) of Zr is almost the same as or greater than that of Hf, while the order of D value was reversed in extraction with HDEHP from 1 M $HClO_4$ ($D_{Hf} > D_{Zr}$). This shows that extraction study of Rf with HDEHP is very interesting.

P06 Extraction behaviors of chloride complexes of Nb and Ta with triisooctyl amine for chemical experiment of dubnium (Db)

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We carried out the solvent extraction of chloride complexes of niobium (Nb) and tantalum (Ta) with triisooctyl amine for chemical experiment of dubnium (Db). It has been reported that the extraction yield of Nb is greater than that of Ta in this system. Radiotracers, ^{95g}Nb and ^{179}Ta, were produced in the ^{nat}Zr(d,xn) and ^{nat}Hf(d,xn) reactions, respectively, using the AVF cyclotron at RIKEN. These radiotracers were chemically isolated by an anion-exchange method. The tracers were preserved in concentrated HCl (stock solution). Aqueous solution of 8−10 M HCl containing the tracers was mixed with an equal volume of TiOA in xylene solution, and the mixture was shaken for fixed time. The radioactivity of each phase was measured by a Ge detector. The results showed that the distribution ratio (D) of Nb was approximately 10^2 times greater than that of Ta. However, the distribution ratio of Ta decreased with the lapse of time from the preparation of the stock solution. To investigate a cause of the behavior, we examined various preparation methods of the tracer. It was found that the distribution ratio and the recovery rate of Ta exhibit various values depending on the preparation methods of the tracer.

P07 Solid-liquid extraction of Nb, Ta with Aliquat 336 resin from hydrofluoric acid solution for chemical experiment of element 105, Db

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We have studied the liquid-liquid extraction behavior of Nb and Ta for the chemical experiment of Db. Because we are planning to use ARCA for Db experiment, an Aliquat 336 resin was prepared and liquid-solid extraction of Nb and Ta with the prepared resin were carried out. Radiotracers of ^{95g}Nb and ^{179}Ta were produced by the proton or deuteron bombardments on ^{nat}Zr and ^{nat}Hf metallic foil targets, respectively, using the AVF cyclotrons at RIKEN and Tohoku University. These radiotracers were chemically isolated by an ion-exchange method. The tracers were dissolved in HF solutions of various concentrations and then were mixed with the Aliquat 336 resin in a syringeless filter tube. Control experiments without the Aliquat 336 resin were also performed to determine initially added radioactivity. After shaking and filtration, each supernatant was separately collected in a sample tube, and was subjected to γ-ray spectrometry to deduce the distribution coefficient (K_d). In the experiment, the K_d values of ^{179}Ta on the 32 wt% Aliquat 336 resin were decreased with increasing HF concentration, while those of ^{95g}Nb showed minimum at 10 M HF. In the presentation, results on a slope analysis to deduce extracted species and the comparison with the results of our previous liquid-liquid extraction will be presented.

P08 Feasibility study to determine of americium activity in plutonium particles by alpha-ray spectrometry

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(JAEA)

We are applying a screening technique with alpha-ray spectrometry to the analysis of particles containing nuclear materials in swipe samples. In the present study, individual plutonium particles with known purification dates prepared from a standard reference material (NBS SRM947) were measured with alpha-ray spectrometry. The obtained (^{241}Am+^{238}Pu)/$^{(239+240)}$Pu activity ratios for the particles with the diameters of more than 1 micrometer were consistent with the reference values within the uncertainties. These results indicate that alpha spectrometry can be applied to the quantitative analysis of ^{241}Am in individual plutonium particles.

P09 A simple ^{40}K-removal water leaching combined with AMP method for detection of low-levels of radiocesium in marine products

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The 2011 Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident resulted in the widespread release of large amounts of radiocesium to sea environments over a wide region in the eastern Japan. For radioactive monitorings, ^{134}Cs (half-life: 2.06 y) and ^{137}Cs (half-life: 30.2 y) have been extensively determined for various marine products over japan after the FDNPP accident. Within 4 years from the accidents, concentrations of radiocesium (particularly for shorter-lived ^{134}Cs) in many marine product samples are gradually decreasing to "below Detectable level" by conventional γ-spectrometry except for samples collected from the coastal/offshore around Fukushima areas. In γ-spectrometry of the marine biological monitoring samples non-destructively or after ashing, a Compton continuum caused by 1461 keV γ-ray of ^{40}K disturbs the determination of low-levels of radionuclides. In the present study, to evaluate low-levels of ^{134}Cs and ^{137}Cs concentrations correctly, we developed a simple ^{40}K removal water leaching treatment and applied these procedures to marine products.

P10 Nationwide survey of atmospheric ^{85}Kr activity concentration in Japan

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(Japan Chemical Analysis Center)

Considerable amount of ^{85}Kr is expected to be discharged into the atmosphere from the Rokkasho nuclear fuel reprocessing plant (RP) once it begins full operation. Prior to the beginning of full operation, a nationwide survey of atmospheric ^{85}Kr has been carried out since 2006. Five areas located at different latitudes were selected for air sampling.

Atmospheric ^{85}Kr activity concentrations in Japan ranged from 1.31 to 1.60 Bq/m^3 (mean ± standard deviation(1 σ):1.45 ± 0.054 Bq/m^3) except for the temporary increases caused by the operation of domestic RPs and the accident of Fukushima Dai-ichi NPP. ^{85}Kr activity concentrations of 2.97 Bq/m^3 at Chiba and 3.95 Bq/m^3 at Sapporo were observed in 2006, which were estimated to be the influence of domestic RPs (Tokai and Rokkasho). Several days after the Fukushima Dai-ichi NPP accident in 2011, ^{85}Kr activity concentration of 17.7 Bq/m^3 was observed at Chiba. It was higher than 10 times of the normal level, and it took 3 weeks to return to normal.

Atmospheric ^{85}Kr activity concentrations reached the maximum in 2007. After that, they showed level or decreasing trend. The yearly discharges (2006-2012) from RPs in Europe decreased compared to those before 2006. From these results, a relationship was confirmed between ^{85}Kr concentration in Japan and the yearly discharges from RPs in Europe. The relatively high range of ^{85}Kr concentration (1.37 - 1.60) in 2014 was estimated to be caused by the increase of the yearly discharge in 2013.

P11 Continuous observation of atmospheric radionuclides at Kawasaki, Japan

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Continuous observations of atmospheric radionuclides were carried out at Kawasaki, Japan. Atmospheric concentrations of natural (^{7}Be, ^{208}Tl, ^{210}Pb, ^{212}Bi, ^{212}Pb, ^{214}Bi, ^{214}Pb, ^{226}Ra) and artificial (^{131}I, ^{134}Cs, ^{137}Cs) radionuclides were observed with aerosol samples collected in Kawasaki, Japan. The seasonal variations of several radionuclides during the period from April 2014 to January 2015 were shown in similar to past variations observed at the present observation site. In order to discuss atmospheric behavior of radon- and thoronprogeny, seasonal variations of radionuclides will be determined continuously.

P12 Development of automatic analysis system of Strontium-90 in environmental samples

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An automatic analysis system was developed to analyze radioactive strontium (^{90}Sr) radioactivity in environmental samples. The analysis method was followed to the manual compiled by the Ministry of Education, Culture, Sports, Science and Technology, to determine low level of ^{90}Sr radioactivity concentration in environmental samples. In this research, the procedure of the Sr analysis before milking procedure of ^{90}Y was automated except for a part of the analysis. The analysis system was consisted by the following three apparatuses. Wet dissolution apparatus was constructed with robot, hotplate, reagent supply pump and monitor. Chemical separation apparatus was constructed with robot, hotplate, pH meter, filter device and reagent supplier. Ion exchange apparatus was constructed with ion exchange column, reagent supplier and monitor. The three systems were controlled by an original program under the LabVIEW. Various kinds of ashed environmental samples were used in performance tests of the automatic system. These tests were successful without system trouble. Finally, Sr recoveries were about 40-70 % to whitebait analyses.

P13 An examination of Ra isotope analysis from large volume of freshwater samples

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(Japan Atomic Energy Agency)

An analytical method of low-level radium isotopes by γ-ray spectrometry with combination of in situ pre-concentration by Powdex resin from large volume of freshwater samples and simple $BaSO_4$ coprecipitation method was examined.

P14 Particle size and chemical form of radioactive aerosols of ^{7}Be

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Radioactive materials were released to the environment due to the Fukushima Daiichi Nuclear Power Plant accident in March of 2011. It is important for us to solve the problems concerning migration and diffusion processes of radioactive materials in the environment. So we tried to estimate the particle size distributions of atmospheric aerosols attached ^{7}Be and the chemical state of atmospheric aerosol particles as a carrier of radioactive materials. Average particle size of ^{7}Be was expected to range from 0.55 to 0.83 μm. We found that there mainly exist F^-, Cl^-, NO_3^- and SO_4^{2-} in aerosol particles in air, as far as anions included are concerned. Average particle size of aerosols including SO_4^{2-} was estimated to be in between 0.55 and 0.83 μm. In addition, it was found that the particle size distribution of aerosols including SO_4^{2-} closely resembles that of ^{7}Be-aerosols. So, we conjecture that sulfate would be the potential transport medium for ^{7}Be.

P15 Advance of pretreatment method using supercritical fluid extraction

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The radioactivity concentrations in the environmental samples are analyzed to confirm whether radioactivity released from the nuclear facilities influence on the environment. The pretreatment of radioactive nuclides of alpha and beta emitters in the environmental samples is performed with acid regents to decompose organic matters and extract target nuclides such as strontium-90, uranium and plutonium. However, the pretreatment has bad influence on workers, facilities and environment. Therefore, we try to develop to the new pretreatment method using supercritical fluid instead of acid regents. In this research, we investigate the extraction yields of stable Sr and ^{238}U into the solvent from soil using the supercritical water oxidation (SCWO) reaction on various conditions such as reaction temperature, pressure and duration. The yields of stable Sr were at most about 60%, while ^{238}U were less than 1%S. In the future, we try to apply addition of catalyst into the SCWO to improve the yield of extraction.

P16 Influence of the radon for environmental radioisotopes measurement

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For the strict measurement of environmental radioisotopes, it is important to evaluate the background of Rn-220 and Rn-222-daughters. The experiments were carried out to estimate Rn-220 and Rn-222-daughters in several laboratories. Also, the backgrounds of liquid scintillation counter and Ge detector were measured under N_2 gas purge and, or not. N_2 gas purge was helpful to reduce the interference by radon daughters.

P17 Attachment Behavior of Fission Products to Aerosol Generated by Heating Metal Salts

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The radioactive nuclei containing fission products (FP's) released from Fukushima Daiichi Nuclear Power Plant accident were spread as aerosol particles. It has been reported that aerosols containing radioactive caesium were generated as non-sea-salt sulfate aerosols, while such a selective generation mechanism has not been revealed. To understand the generation mechanism of radioactive aerosols, a generation system for solid aerosols by heating metal salts has been developed and attachment behaviors of FP's to solid aerosols have been studied in the present work.

Powders of sodium chloride or caesium chloride were heated at various temperatures by electric furnace and solid aerosols were produced. Their size distributions were measured by SMPS. The solid aerosols were injected into a chamber in which spontaneous fission nucleus ^{248}Cm was installed. Radioactive aerosols generated by attaching FP's were collected by polycarbonate filters to be subjected to γ-ray spectrometry and attachment ratios of FP's on solid aerosol particles was estimated.

The attachment ratios do not depend on the total surface area of aerosol particles clearly but the chemical species of aerosols. This result suggests that the chemical interaction plays an important role in generating radioactive aerosols by attachment of FP's on solid aerosol particles.

P18 Development of degradation method for analysis of chlorine-36, iodine-129 and technetium-99 in soil samples

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The safety management for final disposal of spent nuclear fuel will include a search for the behavior of ^{36}Cl, ^{129}I and ^{99}Tc in the biosphere. Behavior of these nuclides in the environment is of great interest because of their high mobility. The pyrohydrolysis is one of the conventional decomposition methods for bulk analysis of chlorine, iodine and technetium in soil samples. However, this method has some disadvantages. In this study, we attempt to construct more reliable and accurate analytical method for ^{36}Cl, ^{129}I and ^{99}Tc in soil samples including degradation process. In this method, the soil sample is treated under subcritical water condition in closed vessels. Furthermore, we examined another degradation solution of tetramethyl ammonium hydroxide under the same condition with water degradation. The isotopic ratios of ^{129}I/^{127}I and ^{36}Cl/Cl in the extracted solution were measured by AMS. ^{99}Tc concentration was determined by triple quadrupole ICP-MS. The analytical result of the degradation method in the case of ^{127}I showed 31% of that obtained by the pyrohydrolysis method. It was 60% for ^{129}I. As for the degradation method using TMAH solution, the result was 81% for ^{127}I and 132% for ^{129}I, respectively.

P19 The study for measurement of Ra-226 through radon progenies by Ge detector

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([1]Kyusyu Environmental Evaluation Association)

Generally, the measurements of Ra-226 in soil by Ge detector are based on the detection of Pb-214 or Bi-214 that reached equilibrium with Ra-226. The sample must be sealed up so that Rn is not lost. The experiment was carried out using the sample container of different materials to evaluate leakage of the Rn. The result showed that Rn will leak through the junction of cap mainly, not through the material itself.

P20 Separation and determination of uranium and thorium series nuclides in environmental samples by solid phase extraction method

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A procedure based on a solid phase extraction method for the separations of U- and Th-series nuclides has been developed. The procedure was performed with stacked UTEVA Resin®, Sr Resin® and DGA Resin® cartridges from Eichrom Technologies. Sample solution (containing uranium isotopes and thorium-series nuclides, 8 M HNO_3 media) was passed through the stacked column at a flow rate of 1-2 mL/min to extract uranium and thorium isotopes, lead isotopes, and actinium isotopes to UTEVA Resin®, Sr Resin®, and DGA Resin®, respectively. Radium isotopes were separated by the stacked column, and uranium and thorium isotopes, lead isotopes, and actinium isotopes sequentially separated and purified by UTEVA Resin®, Sr Resin®, and DGA Resin®, respectively. The radiochemical recoveries were 95% for uranium and thorium isotopes, and almost 100% for radium, lead and actinium isotopes.

P21 Study on uranium in coral skeleton - Isotopic composition and speciation by wavelength dispersive XAFS analyses-

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Age determination of coral sample is generally based on the combination of activity ratios of $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ in the skeleton. Validity of the age determination is confirmed by calculation for initial $^{234}U/^{238}U$ activity ratios. However, inconsistency between calculated initial activity ratios from coral and the ratio of modern seawater has been reported. In this study, chemical speciation of U in coral has been investigated together with $^{234}U/^{238}U$ ratio to serve as an aid to clarify this issue. Based on the results from XRD analysis and observation of skeleton structure by electron microscopes, some kinds of coral samples, which have different ages and species, were analyzed by ICP-MS, EPMA, MC-ICP-MS, and wavelength dispersive XAFS after the appropriate pre-treatment for each analysis. Uranium concentration in calcite sample has inverse correlation with Sr content. It might be due to the transformation of crystal structure from aragonite to calcite with diagenesis. The wavelength dispersive XAFS showed that U existed as hexavalent form for all coral samples. The age-corrected values for $^{234}U/^{238}U$ initial activity ratio were uniform and showed the modern seawater value regardless of crystal structures, ages, and species of coral samples. Thus, the transformation could be occurred without isotopic fractionation for U. As a result of this study, it might be suggested that variability for initial $^{234}U/^{238}U$ activity ratio is not caused by the coral condition such as species and crystal structure.

P22 Preparation of natural radionuclides source using gelatinizer for gamma-ray spectrometry

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Thin disk-shaped sources of natural radionuclides were prepared using chemical reagents, KCl including ^{40}K and Lu_2O_3 including ^{176}Lu, for gamma-ray spectrometry. Each chemical reagent was dissolved and a gelatin was added in the solution. Gelling solutions gelatinized with gelatin were dropped upon filter papers and dried up slowly at low temperatures. Radionuclides on these sources were distributed uniformly by using gelatinizer. Several radionuclides in environmental samples were determined with disk-shaped sources prepared using chemical reagents and gelatinizer. The present method using commercially available chemical reagents is applicable to measurements of various radionuclides concentrated on solid phase extraction disk out of a radiation controlled area.

P23 The radiocesium measurement of edible mushrooms and transfer reduction to a fruit body

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The amount of the radiocesium shifted to a fruit bodies could be reduced from a cultivation culture medium more compared with zeolite element use by using zeolite and potassium compound as a culture medium additive in nameko mushroom cultivation .Also we found that effect on increased productivity can also be expected. Therefore we considered whether we could expect a similar effect in other edible mushrooms.

We understood that a fruit body could reduce quantity of the radiocesium which shifted in comparison with zeolite simple substance use more by using potassium sulfate together with zeolite in a velvet shank, Hatake champignon, the Pleurotus ostreatus like Pholiota nameko.

However, it was admitted the tendency that became slightly higher a shift coefficient adversely without being able to confirm the combination effect with potassium sulfate with other several kinds.

P24 Radioactive cesium decontamination of wild mushrooms

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It has been known that mushrooms accumulate larger amount of radioactive Cs than agricultural products. Since Fukushima Daiichi Nuclear Power Plant accident we have collected over 200 species of wild mushrooms in Miyagi prefecture that locates on the north of Fukushima prefecture. Radioactive Cs concentrations in the mushrooms were measured with highly pure Ge detectors and we have reported concentrations of radioactive Cs in mushrooms. However, the quantities are different from the cooked mushroom. We examined the decontamination of radioactive Cs from the mushroom. More than 65% of radioactive Cs was removed by boiling, and nearly 100% was removed by keeping in salt. K showed the same behavior as radioactive Cs.

P25 Improvement of an Online Perturbed Angular Correlation Measurement System for the Determination of an Electric Quadrupole Moment of the Short-Lived ^{19}F(←^{19}O) Probe

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We have been working on precise measurement of the quadrupole (Q) moment of the second excited state of ^{19}F (I = 5/2) fed through the β^- decay of ^{19}O by means of the perturbed angular correlation (PAC) technique. Because the half-life of ^{19}O is as short as 26.9s, however, it is difficult to introduce the nuclides into samples by usual chemical treatments. Therefore, we have developed an online PAC measurement system coupled to an accelerator, where recoil ^{19}O nuclei produced by the nuclear reaction of ^{18}O(d, p)^{19}O can be implanted kinetically in the sample of interest. For an operation check of the system, we measured cascade γ rays emitted from a standard ^{133}Ba source during beam irradiation. Although background rate became higher as compared with that measured without beam irradiation, the half-life of the intermediate state ($T_{1/2}$=6.27 ns) was successfully observed. Because the level of radioactivity of ^{133}Ba is about the same as the production rate of ^{19}O, it was found that the system can be applied to the measurement of ^{19}F(←^{19}O) probe. However, we have not been able to observe the half-life of the second excited state of ^{19}F ($T_{1/2}$ = 89.3 ns). One of the reasons could be the fact that the recoil ^{19}O nuclei did not come out of the collimator. Improvement of the measurement system is now underway.

P26 Mössbauer spectra and magnetic property of diluted Fe doped Perovskite, $SrSn_{1-x}Fe_xO3\ \delta$

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SrSnO3 doped with dilute magnetic ions has the potential application to spintronic materials. Our purpose is the development of new dilute magnetic at room temperature.

$SrSn_{(1-x)}Fe_xO3$ (x = 0.005 - 0.15) were prepared by using a sol-gel method and heating at 400 °C for 2 hours, 850 °C for 2 hours and 1000 °C for 4 hours or 400 °C for 2 hours, 1000 °C for 2 hours.These oxides were investigated by X-ray diffractionmetry (XRD), a vibrating sample magnetometer (VSM) and Mössbauer spectrometry. Perovskite oxides with less than 60 nm in diameter were obtained and the lattice parameters decreased with the increase of Fe doping rates. Some samples showed the ferromagnetism although any oxides were not so strong. Two doublets and two or four magnetic sextets were observed in the Mössbauer spectra of SrSnO3 doped with more than 8 % Fe. The two or four sextets may be attributed to the Fe^{3+} occupied at the octahedral and tetrahedral sites of ferrites precipitated, respectively. Only paramagnetic doublets were observed for SrSnO3 doped with less than 2 % Fe although the VSM results showed a weak ferromagnetism. This may be due to the induced magnetic defects. Two paramagnetic doublets may be observed by the different configuration between oxygen vacancy and substituted Fe ions.

P27 Measurements of characteristic muonic X-rays using RCNP-MuSIC muon source

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MuSIC, the intense DC muon-beam facility, have been constructed at the Research Center for Nuclear Physics (RCNP) in Osaka University, and a beam commissioning have been performed for starting of research use. We performed preliminary experiments of characteristic muonic X-ray measurement at RCNP-MuSIC.

The experiment was performed at MuSIC-M1 beam line. The copper plate was irradiated by the muon beam decelerated by the aluminum plate, and characteristic muonic X-ray emitted from muonic atom was measured by high purity germanium detector.

The characteristic muonic copper X-rays were successfully detected. Muonic X-ray spectrum for 84 minutes muon irradiation gave a clear μCu (4-3) X-ray peak with 7% statistic error. A good S/N ratio as 5 in μCu (4-3) peak was obtained. In RCNP-MuSIC, additional shield is now constructing to use a stronger incident proton beam, and various application studies using intense muon beam will soon be possible.

P28 Compositional Study of Iron-Containing Mineral in an Ordinary Chondrite Saratov by Mössbauer Spectroscopy

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Chondrites are the most primary source of planetary bodies; compositional studies on mineral containing iron, the main component element of chondrite, are therefore very important for obtaining information on the formation process of our solar system. In this work, we carried out a Mössbauer spectroscopic study on an ordinary chondrite Saratov (L4). A Mössbauer spectrum for Saratov has already been shown by Oshtrakh et al; however, information about magnetism of each mineral is insufficient because the measurement was performed only at room temperature. We thus introduced a low-temperature Mössbauer spectrometer and obtained a spectrum for Saratov at 10 K. The spectrum consists only of components related to primary minerals of chondrite (olivine, pyroxene, FeS, and Fe-Ni), and even at this low temperature, new sextet arising from phase transition of superparamagnetic doublet expected by Oshtrakh et al. was not detected, revealing that Fe^{3+} ions do not to exist in our sample. This observation suggests that the mineral containing Fe^{3+} was oxidized by weathering on the earth after its falling on the ground.

P29 Characterization of the stable isomers for U(IV) and U(V) complexes with Schiff-base ligands

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For characterization of stable isomers for eight coordinate U(IV) and U(V) with Schiff-base ligands, DFT calculations were performed to square-antiprismatic U(IV) and U(V) complexes and to dodecahedral U(IV) and U(V) complexes. In the geometrically optimized structures of the complexes, the U-O and U-N bond distances were similar to those determined by single crystal X-ray diffraction analysis. The total bonding energies for the U(IV) isomers indicate that the more stable isomer is the complex having a square-antiprismatic structure. The square-antiprim U(IV) is 7~8 kJ/mol more stable compared with the dodecahedral structures. In the case of U(V), the total bonding energies of the isomers revealed that the dodecahedral structure is more stable than the square-antiprismatic structure. The results by DFT calculations are agreed well with the results obtained from experiments.

P30 Investigation of the process for aggregate formation with the reduction of uranium

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It is important to understand the behavior of actinide ion in nature environment. In previous study, it was found that aggregates of U(IV) formed in reduction process of U(V) enhanced the rate of the disproportionation of U(V) and the reduction current of U(V). In this study, we investigated the relationship between reduction process and formation of aggregates by measuring electrochemical quartz crystal microbalance (EQCM). When EQCM measurements of the reduction of U(V) to U(IV) in weakly acid solution were performed, the frequency was changed negatively. This indicates that aggregates of U(IV) were formed on the electrode. We consider that aggregates form in the three steps from the deposition rate; the induction step (~start deposition), early stage aggregation step (deposition amount ~4 nmol) and secondary stage aggregation step (deposition amount >4 nmol). The pH dependence of the reduction and deposition rate was investigated. The U(IV) aggregates formed at more than pH 3.1. When pH was higher, the induction time was fast, and aggregation rates at early and secondary stage were high. This suggests that a U hydroxide relates to the formation of aggregates.

P31 Selective extraction of cesium and strontium

～solvent extraction III by risk-free solvent～

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High level nuclear waste includes ^{137}Cs and ^{90}Sr, both are one of the most important long-lived fission products. Recently solvent extraction has been studied in order to remove these nuclides from the high level nuclear waste. The solvent extractions are one of the most useful separation methods of metal ions. Using 1,2-dichloroethane, 1-octanol or chloroform as a diluent and crown ethers can functions as extractants of cesium and strontium from acid solutions[1][2]. The diluents have risks of fires, explosions and environmental soundness. In our previous works, the solvent extraction of uranium from nitric acid solution was conducted by HFC-43-10mee（DuPont; Vertrel XF）-TBP system. The diluents show excellence in the noninflammability, the low toxicity, and chemical stability.

In this study, we investigated the use of azeotrope of HFC-43-10mee as a diluent. We will present the results alkali metals and alkaline earth metals from acid solution by dicyclohexano-18-crown-6-ether as an extractant in this diluent.

[1] R. A. Sachleben, et al., Sep. Sci. Technol., 32(1997)275.
[2] K. K. Gupta, et al., Sol. Extr. Ion Exch., 21(2003)53.

P32 Synthesis of mono-triazinyl-phenanthroline derivatives for MA/Ln partitioning and the performance evaluation in lanthanide series elements.

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It is one of the essential issue to separate Minor Actinides (MA) and lanthanides from high level radioactive waste from the aspect of Partition & Separation (P&T). However, the practical/appropriate methods have not been constructed. In this study, we focused on solvent extraction using 2-(1,2,4-triazine-3-yl) 1,10-phenanthroline (t-Phen) which may contain tridentate coordination site. The performance evaluation such ligands are hardly investigated.

T-Phen ligands, having different substituents (methyl group, ethyl group, phenyl group) at 5,6-position of each triazine ring, were synthesized from 1,10-phenanthroline-2-carboxamidehydrazone with α-diketone following known procedures. All the ligands were obtained as hydrates identified by ^{1}HNMR and TG/DTA. The extraction experiments of lanthanide series elements except for promethium were performed with changing the nitric acid concentration, the ligand concentration and the organic phase compositions. The concentrations of lanthanides in aqueous phases were measured by ICP-MS. UV-Vis spectroscopic titrations for t-Phen with Eu^{3+} or H^+ in acetonitrile solution was also performed. Other details for substituent effect for three ligands will be presented in our poster.

P33 Structure and stability of EDTMP and DOTMP complex with Th(IV) for nuclear medicine

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In our experiments where Th-227 labeled EDTMP was administered to mice, the selective accumulation of daughter nuclides Ra-223 and Rb-221 to bones, and the clearance from non-target organs have been found. On the basis of this result showing that Th-EDTMP is collected by bones at high ratio, we are studying the complexes of EDTMP and DOTMP with alpha-ray emitting nuclides.

In this study the stability constants of the complexes of EDTMP and DOTMP with Th(IV) was determined. In the titration experiments using a series of metal ions, DOTMP shows larger l logK_{ML} values for d transition metals, because the number of nitrogen donor atoms are larger by two in DOTMP than EDTMP. In contrast to d transition metals, Th shows similar behaviors to Ca, rather than to d transition metals, in values of logK_{ML} and logK_{MHL}. This is indicative that the coordination by nitrogen donor atoms is rather weak.

Also, the bond lengths between Th and donor atoms were elucidate by crystallography. In the DOTMP complex, the coordination is much stronger than in the EDTMP complexes. However, Th-N bond length is the longest among the known complexes.

P34 Production of ^{67}Cu for cancer therapy with accelerator neutrons by deuterons

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([1] Japan Atomic Energy Agency, [2] Chiyoda Technol Corporation)

Copper-67 is an attractive radionuclide for cancer therapy, because of emissions of medium energy beta particle (a mean energy of 141 keV) and gamma rays (91, 93, 185 keV) suitable for imaging, and its appropriate half-life (62 hours). However, the use of ^{67}Cu in clinical research has been limited due to the difficulty in obtaining a sufficient amount of ^{67}Cu. In this study, we have investigated the production of ^{67}Cu by the ^{68}Zn(n,x)^{67}Cu reaction ($x=n'p,\ d$) using fast neutrons from ^{nat}C(d,n) or ^{nat}Be(d,n) reaction. The highly purified ^{67}Cu was obtained from a considerable amount (33 g) of ZnO sample by two columns (chelate resin chelex-100 and anion exchange resin AG1-X8). By using the obtained ^{67}Cu solution, the labelling of DOTA and TETA which are useful bifunctional ligands for the labelling monoclonal antibodies was succeeded in more than 97% yield. In addition, we successfully developed a recycling process of the used ^{68}ZnO sample by precipitating ^{68}Zn(OH)$_2$ from the separated Zn fraction. It was confirmed that the recycled sample did not include the extra impurities inhibiting the labelling reaction of DOTA or TETA.

P35 Recovery and purification of ^{99m}Tc isolated from ^{99}MoO$_3$ using a thermal separation technique

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^{99m}Tc is a daughter nuclide of ^{99}Mo and is the most widely used radionuclide in nuclear medicine diagnosis. The thermo-separation of ^{99m}Tc from ^{99}Mo produced using accelerator neutrons has been found to be a promising method to obtain a high purity ^{99m}Tc from ^{99}Mo offering a constant high efficiency. Irradiated ^{100}MoO$_3$ samples are heated in a tube furnace where ^{99m}Tc alone is thermally separated from the samples. Post-separation procedures involved washing the deposited ^{99m}Tc from the quartz tube wall in the low temperature region. The collected ^{99m}Tc was loaded onto an alumina column which is then eluted with a small quantity of saline solution to obtain ^{99m}Tc as TcO$_4^-$. This was previously performed with 0.1 mM NaOH solution to allow ^{99m}Tc adsorption onto an alumina column (SepPak Alumina N Plus Light, Waters), however the washing efficiency was erratic (50-90%) affecting the total yield. In this study the 0.1 mM concentration of NaOH was substituted by 0.1-0.05 M to increase the solubility of ^{99m}Tc in the solution, which was then neutralized by a cation-exchange column (Dionex OnGuard II-H, Thermo Fisher Scientific, Inc.) coupled with an alumina column for ^{99m}Tc adsorption. As a result, a high ^{99m}Tc washing efficiency (> 95%) was consistently obtained leading to a high total ^{99m}Tc yield using the thermo-separation system.

P36 Extraction experiment in a closed system using syringes for a simulation of ^{211}Rn/^{211}At generator

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The ^{211}At isotope ($T_{1/2}$= *7.2* h), an α emitter, has attracted attention as a nuclear medicine for the treatment of cancer. Because α-particles are expected to make higher therapeutic effect than β-particles due to high linear energy transfer. There is also a feature that can be treated without damaging the normal cells around a tumor. The half-life of ^{211}At is, however, not long enough to use in areas remote from the production facilities. We produced ^{211}Rn ($T_{1/2}$= *14.6* h) in the reaction of ^{209}Bi(^{7}Li,5n)^{211}Rn using the JAEA-Tokai tandem accelerator to study the establishment of a generator technology that enables the remote location use by milking an ^{211}At isotope as a daughter nuclide of the generator. In this study, we devised a generator of a complete closed system by using two syringes, and performed a trial for milking of ^{211}At from ^{211}Rn by solvent extraction.

P37 Production of Radioisotopes for Nuclear Medicine Diagnosis and Therapy at RCNP Osaka University

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Today, the use of different RI with the diversification of the nuclear medicine diagnostics and therapy have been made. Almost all of the ^{99m}Tc that are used in SPECT rely on imports, but has been studied much be produced using domestic accelerators. Also in the PET diagnostic ^{18}F but is being used to keep a number of facilities at home and abroad, the use of β+ emitting nuclides of long life has been demanded. Furthermore lest therapy with α-ray has also been studied, ^{211}At has been taken up as its promising nuclide.

We have produced the following RI for nuclear medicine to date using the AVF cyclotron and ring cyclotron at RCNP. The ^{52}Fe ($T_{1/2}$ = 8.28 h), ^{62}Zn ($T_{1/2}$ = 9.26 h), ^{65}Zn ($T_{1/2}$ = 244.1 d), ^{99}Mo ($T_{1/2}$ = 65.94 h), ^{99m}Tc ($T_{1/2}$ = 6.01 h), ^{124}I ($T^{1/2}$ = 4.18 d), ^{191}Pt ($T_{1/2}$ = 2.91 d), ^{210}At ($T_{1/2}$ = 8.14 h), and ^{211}At ($T_{1/2}$ = 7.21 h) were prepared.

P38 Production of medical radioisotope ^{90}Y at a zirconium target irradiated by fast neutrons in the ^{9}Be(p,*n*) reaction

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Today in nuclear medicine, many kinds of radioisotopes play an important role in radiological diagnosis and therapy of a specific disease such a cancer. In particular, for yttrium-90 it is well-known that the^{90}Y-labeled radiopharmaceutical such as ibritumomab tiuxetan (ZEVALIN®) are widely used for the treatment of non-Hodgkin’s lymphoma (NHL) in the world. We have ever investigated the excitation function of^{90m}Y associated with the production of^{90}Y on a zirconium target by a fast neutron beam, and also estimated the optimized production condition of the^{90m}Y nuclides with respect to primary proton-induced beam energies, target condition and productions of by-products (^{89}Zr,^{87m}Sr and so on). Each of fast neutron beams was generated along with 15 to 30 MeV proton-induced reactions on a beryllium target using a cyclotron accelerator such a medical PET cyclotron currently in use. The purpose of this work is to reproduce distributions relative to fast neutron fluxes around the target position along with each of the primary proton-induced energies using the Monte Carlo simulation code of PHITS, and also to estimate the productions of^{90m}Y and those by- products on the assumptions of various source and tally conditions based on our developed production reaction system using the DCHAIN-SP2014 program included into the PHITS code package.

P39 Lateral distribution and seasonal variation of ^{228}Th/^{228}Ra ratio in surface seawaters of the East China Sea and the Sea of Japan

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A total of ~100 surface seawater samples were collected with in the Sea of Japan and the East China Sea (ECS) in 2008-2014, and lateral and seasonal variations of ^{228}Th ($t_{1/2}$ = 19.1 y)/^{228}Ra ($t_{1/2}$ = 5.75 y) ratio were examined by low-background gamma-spectrometry. In early summer, within the ECS, the ^{228}Th/^{228}Ra ratio of the continental shelf water-rich surface waters was markedly lower relative to the Kuroshio water, being due to preferentially removal of ^{228}Th from the surface by a high flux of biogenic particles. The ^{228}Th/^{228}Ra ratio at surface in the Sea of Japan exhibited small variation with the range of the eastern ECS. Combining with the seasonal variation of ^{228}Th/^{228}Ra ratio and short-lived ^{234}Th ($t_{1/2}$ = 24 d), we discuss the scavenging mechanisms and behaviors of reactive components at surface within the Sea of Japan.

P40 Leaching of uranium and thorium series nuclides from pitchblende

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The leaching behavior of Ra isotope from pitchblende, radioactive mineral, was observed along pH of HCl aqueous solutions in contact. The activity ratios of ^{223}Ra/^{226}Ra in the leachates were observed to be larger than that in the mineral and tended to increase with an increase in pH of the HCl aqueous solutions. This tendency indicates that ^{223}Ra is preferentially leached as compared with ^{226}Ra. Radium-226 and ^{223}Ra are formed through three alpha decays from the parent nuclides which ^{238}U and ^{235}U, respectively. The extent of damage around a nuclide is assumed to be proportional to the total alpha energy released in the formation of the nuclide. The summation of all alpha energy released in transformations from ^{235}U to ^{223}Ra is larger than that in the transformations from ^{238}U to ^{226}Ra. Therefore, the difference in leaching efficiency of ^{223}Ra and ^{226}Ra was estimated to be due to the difference in total alpha energy released in the formation of the nuclide.

P42 Cross section measurement of 80 MeV neutron induced reaction for meteorite constituent elements

NINOMIYA, K.[1], NANBU, A.[2], SHIGEKAWA, Y.[1], TAKAHASHI, N.[1], SHINOHARA, A.[1], SEKIMOTO, S.[3], YASHIMA, H.[3], SHIMA, T.[4], HAGIWARA, M.[5], IWAMOTO, Y.[6], SHIBATA, S.[7], CAFFEE, M. W.[8], NISHIIZUMI, K.[9]

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The determination of cross-sections by neutron-induced nuclear reaction is very important in the view point of space and planetary sciences to investigate the history of cosmic ray exposure. These data are also required for estimation of residual radioactivities in accelerator facilities. However, cross section measurements in the energy region have scarcely been limited due to difficulty in obtaining high energy monoenergetic neutrons. Our group has developed a method to extract the component of high energy monoenergetic neutrons at the Research Center for Nuclear Physics (RCNP), Osaka University. This method is based on two neutron irradiation experiments produced by Li (p, n) reaction with two different angles for the axis of the primary proton beam to correct the contribution of the low energy neutron reaction. In this study, high-energy monoenergetic neutron-induced reactions with 80 MeV have been studied for meteorite constituent elements, and cross sections of short-lived radionuclides were determined by gamma-ray spectrometry.

P42 Seasonal variation of radioactive cesium concentration in Fukushima City

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A large amount of radioactive nuclides were released in the environment by the accident at the Fukushima Daiichi Nuclear Power Plants (FDNPP) in March 2011. We have been collecting air-dust on a filter using high volume air sampler at Fukushima city (Fukushima Pref.), Marumori town (Miyagi Pref.) and Hitachi city (Ibaraki Pref.) just after the accident. We identified the radioactivities of ^{134}Cs and ^{137}Cs in the filters using HPGe detector. In this paper, time variation among three sampling point and air-dust size distribution of radioactive concentrations will be discussed.

P43 Measurement of radioactive Cs in monkey cartilages collected in Minamisoma city

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

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We determined Cs-137 concentrations of cartilages and bones in monkeys that inhabited in Minamisoma City. The concentrations of cartilages and bones were 6631±14 Bq/kg and 3725±14.46 Bq/kg in young monkey and 415.4±5.16 Bq/kg and 178.4±1.94 Bq/kg in child monkey. The concentrations of cartilages were higher than that of bones.

Stable Cs concentration of cartilages and bones were also determined to be 769±37 ppb and 373±19 ppb in young monkey and 47.3±1.4 ppb and 29.3±1.0 ppb in child monkey. The stable Cs concentrations of cartilages were higher than that of bones the same as the Cs-137 concentrations. Specific activities of Cs-137 of cartilages and bones were 9.45±0.24 Bq/μg Cs and 8.67±0.36 Bq/μg Cs in young monkey and 10.1±0.3 Bq/μg Cs and 6.70±0.24 Bq/μg Cs in child monkey.

The specific activities of Cs-137 were constant between the cartilages and bones. The result showed that the differences of the Cs-137 concentrations between the cartilages and bones don't depend on the Cs-137 pollution at the resided area of monkeys. The cartilages contained large amount of Cs-137 because the cartilages contained more stable Cs than the bones and Cs-137 incorporated into cartilages with stable Cs.

P44 Evaluation of Influence of the Fukushima Dai-ichi Nuclear Accident on Environmental γ -ray Dose Rate in Miyagi Prefecture

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In this paper, evaluation of radiocesium (Cs-134+137)-contribution in air-absorbed γ-ray dose rate observed in Miyagi Pref., adjacent to Fukushima Pref., was discussed in order to estimate the influence of the Fukushima Dai-ichi Nuclear Accident. Environmental γ-ray spectra were measured in Miyagi Pref. by using a 3”φ×3” NaI(Tl) detector and the data were processed by a G(E)-function method and a 49×49 response matrix (RM)-method, which was specially developed to evaluate radiocesium dose contribution. In evaluating radiocesium contribution, the RM-method needs only one-time spectrum measurement, though the G(E)-function method needs natural background dose-rate data before contaminated by radiocesiums from the Fukushima Dai-ichi Nuclear Accident. In our study, about 20-100 nGy/h in total 40-140 nGy/h were evaluated as radiocesium dose contribution. Total and radiocesium dose rates obtained by the RM-method agreed with those of the G(E)-function method, within 5% and 15%, respectively.

P45 Development of the aerosol generation system for simulating the dry deposition behavior of radioaerosol emitted by the accident of FDNPP

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A large amount of radioactivity was discharged by the accident of FDNPP. The long half-life radionuclide, ^{137}Cs was transported through the atmosphere mainly as the aerosol form and deposited to the forests in Fukushima prefecture. For evaluation of behavior of ^{137}Cs in vegetation system, fundamental data for deposition and uptake process of sulfate aerosol was desired. In this study, we developed aerosol generation system for simulating the dry deposition and the foliar uptake behaviors of aerosol in the different chemical constitutions. In this system, the method of aerosol generation based on the spray drying. Solution was send to a nozzle by a syringe pump and spraying with a high speed air flow. The sprayed mist was generated in a chamber in the relatively high temperature. The solution in the mist was dried quickly, and micro size solid aerosols were generated. The experimental condition, such as the size of chamber, chamber temperature, solution flow rate, air flow rate and so on, were optimized. In the presentation, we will discuss the performance of the aerosol generation system.

P46 Vertical distribution of radioactive cesium in the sediments of Tokyo-bay and Chiba’s urban river released by the Fukushima Dai-ichi nuclear power plant accident

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To make clear the behaviors of radioactive cesium in hydrosphere released by the FDNPP accident, we measured its vertical distribution in the two groups of sediments, in Tokyo-bay located off Makuhari and in Chiba’s urban river including Hanami-river and Shinkawa. For analysis in Tokyo-bay, two types of sediments (an artificial deeper site and a natural shallower site) were compared from February ’13 to August ’13. Radioactive Cs in both sediments had gone deeper and deeper, reaching 40 cm layer in this area, while more than 90 % of Cs in soil is recognized to stay in the upper 5 cm layer. In addition, the inventory of Cs had increased. The phenomenon was especially observed more clearly at the deeper site than at the shallower site. It is supposed that the deeper site in Tokyo-bay have the feature of collecting Cs. On the other hand, about the urban river, Cs had also gone to deeper sediments in some areas, reaching about 30 cm to 50 cm layers. Moreover, the concentration of Cs in each layer of the river's sediments is clearly higher than Tokyo-bay's. It is suggested that urban rivers behave not only as Cs-carrier to sea but also as Cs-storage in themselves.

P47 Rapid Determination Method for ^{90}Sr in Seawater from ^{90}Y Measurement Through Yttrium Phosphate Coprecipitation and Purification with DGA Resin

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Strontium 90 is one of the nuclides that have been released to the environment by the Fukushima Daiichi nuclear power plant accident. The radioisotope has been leaking to the underground and released into the ocean with contaminated water. The rapid measurement of ^{90}Sr in seawater is necessary for environmental monitoring in Fukushima. It requires separation and purification of either of ^{90}Sr and ^{90}Y. We have been engaged in examination to improve the procedures of radioactive Sr analysis. In this study, Sr and Y were separated through yttrium phosphate coprecipitation. Y was purified through DGA resin solid phase extraction and DDTC solvent extraction. Radioactivity of ^{89}Sr can be estimated by using combination of measured radioactivities of Sr (^{89}Sr + ^{90}Sr) using Sr Rad-Disk method and ^{90}Sr by ^{90}Y measurement.

P48 Dispersion of radioactive cesium released from the Fukushima Dai-ichi NPP in coastal area off Fukushima Prefecture

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Nuclear accident occurred on 11 March 2011 from stopping the cooling system and power supply at the Fukushima Dai-ichi Nuclear Power Plant (NPP) by the Great East Japan Earthquake and tsunami. Radioactive cesium (^{134}Cs, ^{137}Cs) was released from the NPP into the environment. The dispersion of radiocesium added to the ocean is essential to understand the impact of radiocesium to the coastal ocean off the Fukuhisma Prefecture. Therefore we determined the spatial and temporal variations of radiocesium in the Fukushima coastal areas and evaluated the horizontal and vertical variations by measuring the concentration of radioactive cesium in the periodic sampling of coastal seawater at Iwaki off the coast by the Fukushima Marine Science Museum Foundation, and the seawater that was collected by Tansei-Maru research expeditions in August 2012 (KT-22-12) and in January 2013 (KT-13-01).^{134}Cs concentration of the KT-13-01 samples showed an approximately 200-times higher value at the maximum (in the case of comparing the C03 and J10) than those of the KT-12-22 samples. The results of periodic sampling will be also used for discussing the situation of diffusion behavior of radiocesium.

P49 Environmental dynamics of dissolved

radionuclides and ions in riverine water flowing urban area after the FDNPP accident

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To observe the fate/dynamics of radionuclides, which has been dispersed from FDNPP, in river water flowing through densely-populated area, we investigated the concentration of dissolved radiocesium ^{137}Cs and radioiodine ^{129}I and their stable isotopes (^{133}Cs and ^{127}I) in riverine water, Japan. Water samples were collected at 6 stations in the Ohori River system, Chiba (about 200 km away from the FDNPP). Additional two inflow waters to this river were sampled at the same time. These samples were filtrated with membrane filter (pore size 0.20 μm), and the radioactivity of ^{137}Cs in the filtrated water was determined by HP-Ge gamma-spectrometry. Dissolved ^{129}I was measured by accelerator mass spectrometry (AMS) at MALT, The University of Tokyo after the appropriate treatment of samples. Other dissolved elements were determined by ICP-MS. The average dissolved ^{137}Cs radioactivity decreased by 77% through 2012 to 2015 in this river. The concentration for ^{137}Cs could be affected by the inflow to this river, that is, the concentration for ^{137}Cs was diluted with input water which has lower concentration compared with that of the Ohori River. ^{129}I and major cation (Ca^{2+} and Mg^{2+}) concentrations in soluble fraction showed the good correlation with those of ^{137}Cs.

P50 Simple characterization of radiocesium in sand sample combined with the imaging plate

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A sand sample collected at the sandbox in a park at Fukushima was selected to know the behavior of radiocesium. Fukushima-derived radiocesium (^{137}Cs) in the sand sample were analyzed by a sequential extraction procedure. The radioactive cesium isotopes were divided into five fractions by the procedure based on Tessier's method. The characterization of radioactive cesium in the sample was achieved by γ-ray spectrometry for each extraction residues. About 66% of the radiocesium existed in free oxides, organic-bound and residual material. To acquire a two-dimensional image of sand particle, radioactive materials in the sieved sand sample were measured using the imaging plate for 1 day. The radioactive materials were distributed spotty, suggesting that the number of radioactive particles was relatively small but that their activity levels were relatively high. The result indicates that radiocesium in sand sample will have been retention to soil particle.

P51 Temporal changes in atmospheric ^{137}Cs concentrations in Namie Town, Fukushima

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The temporal changes of atmospheric ^{137}Cs concentration in the area with relatively high contamination by the Fukushima Dai-ichi Nuclear Power Plant accident were investigated to reveal the factors affecting the concentration of re-suspended ^{137}Cs in the air. Aerosol samples with larger and smaller than 1.1 μm diameter were separately collected during one month using a high volume air sampler at the Minami-Tsushima District in the western area of Namie Town during October 2012 to December 2014. The observed total atmospheric ^{137}Cs concentrations ranged 0.042-1.1 mBq m^{-3} and showed clear seasonal fluctuation with high ^{137}Cs concentration during summer-autumn and low during winter-spring. Most of atmospheric ^{137}Cs was contained in larger particles than 1.1 μm during summer-autumn, suggesting that fluctuation of large particles is related to the seasonal change in the total atmospheric ^{137}Cs concentration in this area. During the high and low atmospheric ^{137}Cs concentration interval, easterly and westerly winds were dominant, respectively. The ^{137}Cs inventory on the ground was reported as relatively high in east direction from the measurement site and low in west. Those results suggest that the atmospheric ^{137}Cs concentration at the measurement site was strongly influenced by larger particles from the area with relatively high contamination.

This work was performed under contract with the government of Aomori Prefecture, Japan.

P52 Dosimetry using milk teeth of Japanese children by measuring CO_2 radical

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The releases of radioactive materials from the nuclear plant and/or from the military program activities result in an ionization radiation exposure to animals and people, the precise dose assessment is required on the point of view of the radiation protection, radiation safety, and so on. Electron spin resonance (ESR) method is one of a powerful tool that has been used in the study of dosimetry of animals, especially measuring CO_2 radicals of teeth which produced by the exposure of the radioactive materials and UV light. In this work, enamel of the milk teeth of Japanese children were isolated from dentine and crashed into a grain, and then irradiated with 60Co gamma-ray in air at room temperature with an absorbed dose up to 1 Gy. From the dose response curve, the detection limit of our ESR system was estimated to be 34 mGy (+- 16 mGy).

A part of this study is the result of "Establishment of the dosimetry protocol using milk teeth of children" carried out under the Strategic Promotion Program for Basic Nuclear Research by the Ministry of Education, Culture, Sports, Science and Technology of Japan.

P53 Distribution of radiocaesium in the wild Japanese monkey (*Macaca fuscata*) after the accident of Fukushima Daiichi Nuclear Power Plant.

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We have set up a study group for the assessment of radiation effects of the accident of Fukushima Daiichi Nuclear Power Plant. In this study, we collected 18 organs (femoral muscle, longissimus muscle, masseter muscle, tongue, diaphragm, heart, lung, liver, spleen, kidney, pancreas, bladder, testis, brain, adrenal gland, submandibular gland, thyroid gland and thymus gland), peripheral blood, bile and urine from wild Japanese monkey (*Macaca fuscata*) in Fukushima prefecture and determined the activity concentration of radionuclides in these organs. The deposition of only ^{134}Cs and ^{137}Cs was observed in all the organs examined. The radioactive caesium level in the skeletal muscle was the highest and that in the thyroid gland was lowest among all the organs studied. The mean radiocaesium concentration of skeletal muscle in monkeys captured in Fukushima was 3,066.5 +/- 7,911.6 Bq/kg, in the range of 61.6 +/- 1.5 to 82,586.4 +/- 75.5 Bq/kg. There is no great difference in the radiocaesium concentration of skeletal muscle in each monkey which captured in same area. Based on these results, we are now estimating the dose of internal and external exposure to radiocaesium in monkeys in Fukushima.

P54 ^{137}Cs and ^{129}I concentrations in seawater off Fukushima Prefecture in 2013-2014

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Concentrations of ^{134}Cs, ^{137}Cs and ^{129}I in seawater samples collected off Fukushima during the R/V Umitaka-Maru UM-13-05 cruise in May 2013 and R/V Sinnsei-Maru KS-14-6 cruise in May 2014, after the Fukushima Daiichi Nuclear Power Plant (F1NPP) accident, were measured. For concentrations of ^{137}Cs and ^{129}I in surface seawater at 5km away from F1NPP in 2013-2014, ^{137}Cs was reduced from 36.8 mBq/kg to 12.5 mBq/kg and ^{129}I was reduced from 1124nBq/kg to 307nBq/kg. The ^{129}I/^{137}Cs ratio did not change between 2013and 2014.

P55 Examination of the radiocesium excretion effect by the lactic acid bacteria

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For the purpose of the removal of ^{137}Cs from the human body, the adsorption ability of the lactic acid bacterium preparation for ^{137}Cs was examined and evaluated. Lactic acid bacterium preparation and ^{137}Cs (ca.1 kBq) were introduced into water, pH 6.8 fluid and pH 1.2 fluid, respectively, and shaken. After 1-6 hours, the ^{137}Cs adsorption rate of the preparation reached the maximum value, that is, 46% in water, 21% in pH6.8 fluid, 17% in pH1.2 fluid. By adding potassium to the lactic acid bacterium preparation, until 20 mmol L^{-1} the ^{137}Cs adsorption rate was reduced. However, above this concentration, the ^{137}Cs adsorption rate was almost unchanged.

These results show the lactic acid bacterium preparation have adsorption ability for ^{137}Cs in stomach and intestines. Therefore, the proposed method will be one of the effective means to prevent radiation emergency by ^{137}Cs.

P56 Syntheses of Novel Organic Composite Adsorbents and Its Application for Simultaneous Separation of Various Types of Radioactive Nuclides in Seawater

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Triggered by the great Tohoku earthquake and tsunami on March 11, 2011, an enormous amount of contaminated water containing radioactive nuclides such as cesium, strontium, and iodine was generated by means of sustained injections of water or seawater for emergency cooling of nuclear cores in Fukushima Daiichi Nuclear Power Station. Thus, promising adsorbents for effective separation of various types of radioactive nuclides under various environments have been required for nuclear severe accidents. Based on this viewpoint, novel tannic acid-type resin combined with strongly basic anion-exchange resin, named improved tannic acid-type resin (ITA-02 resin) was synthesized successfully and the adsorption behavior of various types of radioactive nuclides such as cesium, strontium, lanthanides, actinides, and iodine on ITA-02 resin have been studied in several types of seawater at a temperature range of 278 - 318 K, compared with those of some typical resins. As a result, it was found that ITA-02 resin can adsorb strongly these species in seawater at room temperature. From these results, we have proposed that ITA-02 resin is suitable for selective separation of typical radioactive nuclides in contaminated seawater containing radioactive nuclides from nuclear severe accidents.

P57 Development and Application of sequential separation technique for ultra-trace multi-elements including Pu

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Abundance and isotopic ratios of trace uranium, thorium, lead and lanthanides in environmental samples play a key role to investigate features of the samples. Plutonium is an artificial element which originates from nuclear bomb tests and accidental releases from nuclear power plants. The authors accomplished the development of technique for sequential separation of U, Th, Pb, the lanthanides, and Pu using a single anion-exchange column and mixed media consisting of hydrochloric acid, nitric acid, acetic acid, and hydrofluoric acid. An automatic system assembled for this work sequentially separated the elements of interest for 6.5 h. This automatic separation system (CASSUAL) was served for the elemental and isotopic analyses of ultra-trace elements in the tree ring samples.

P58 Trial production for gamma-ray emitting source applied by SLA 3D printer.

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When the radioactivity in environmental samples was quantified by gamma-ray spectrometry, geometric conditions of the sample greatly affect the measurement.

There is a very valuable sample in the environmental samples. Therefore, it cannot be measured by physical destruction. And so, it was attempted that the dummy gamma-ray source is manufactured by using a 3D scanner and stereolithography apparatus (SLA) type 3D printer.

As a result, with the use of a reagent involving natural radionuclide, it was possible to create a detection efficiency curve to match the shape of the dummy gamma-ray source. Therefore, it has become possible to create dummy gamma-ray source with high accuracy in a short period of time.

P59 Radiation degradation of cable coatings examined by positron annihilation lifetime spectroscopy

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Polyethylene is widely used for covering cables in radiation environments. The radiation degradation of polyethylene is important for the safety of nuclear power plants and accelerators. Nanostructure change of such polymers is required to investigate because it correlates with the macroscopic structure and sample properties. In this work, we measured the change of nanostructure (free volume) by positron annihilation lifetime spectroscopy (PALS).

High density polyethylene and low density polyethylene were used. Electron beam was irradiated up to 1000 kGy under vacuum at room temperature. After the irradiation, samples were exposed and stored in air. The change of the free volume and the chemical structure were measured by PALS and micro-Fourier transform infrared (micro-FT-IR) spectroscopy.

The results of PALS suggest that there are two type of free volume in polyethylene, which were crystalline-amorphous region and amorphous region. Free volume size was unchanged during the long-term storage but positron annihilation relative intensity changed. In HDPE, positron annihilation relative intensities of the crystalline-amorphous intermediate region decreased during long-term storage, whereas that of the amorphous region was unchanged. On the other hand, micro-FT-IR results show that the amount of carbonyl groups increased during long-term storage in particular for HDPE. These results suggest that the different changes of positron annihilation relative intensities in two regions are probably due to the difference of the mobility of the oxidized polymer chain.

P60 Positron source for a positron annihilation lifetime measurement cell under an electric field

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A positron is an antiparticle of electron, and when it’s bound with an electron, it generates a positronium (Ps). The kinetic energy of Ps depends on that of positron, which is just generated. Our goal is to develop a low-energy Ps source by applying the electric field to high-pressure gas. We made a positron source by dripping ^{22}NaCl in a metal rod (2 mm in diameter, 17 cm in length). We use a cylindrical electrode, and attach the positron source in the center. Ps formation amount in the gas is secured, and positron can efficiently accelerate. We estimates positron annihilation ratio in the electrode by positron annihilation lifetime measurement, by which we measure the positron lifetime by the time difference between when a positron injects and when it is annihilated with an electron. We also investigate the Doppler broadening of the annihilation gamma ray in the electrode by germanium semiconductor detector.

P61 Structures of positronic and muonic alkali atoms

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Positron, which is an anti-particle of electron, and positive muon, which has about 207 times heavier mass, have been used as unique probes and tracers for material science and atomic/molecular physics. The positron represents a large quantum effect and delocalizes around atoms. Since annihilation gamma rays, emitted when the positron encounters an electron, reflect the electron momentum, the positron has been a sensitive probe for widely distributed electrons with low momentums. The positive muon has about 2.2 μs and emits a positron. The behavior is like a hydrogen nucleus, but the quantum distribution rised from the smaller mass than the hydrogen nucleus is crutial to the resolution of muon analysis, which utilizes the large magnetic moment of the muon. In this work, we calculated quantum distributions and wavefunction structures of bound and resonance states of positronic and muonic alkali atoms. These systems can be general models for interactions between a positron/positive muon and an atom because we can describe precisely positron/muon active reactions with the valence elecron and static interactions with inner shell electrons. Radial distribution functions and structures of wavefunctions of the positron/muon are revealed by non-adiabatic three-body calculation. This work is a fundamental investigation for positron/muon probe analysis.

P62 Age-momentum correlation measurement of Ps thermalization process in Argon gas

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Positronium (Ps) consisting of a positron and an electron is the lightest atom. Recently, demand for the slow Ps is further increasing, due to developing of the Ps experimental such as production of anti-hydrogen, QED test and Ps BEC. In this study, we investigated the Ps thermalization process in Argon gas by the Age-momentum correlation measurement in which positron annihilation lifetime spectroscopy and time dependence of Doppler broadening (DB) of annihilation photon are simultaneously obtained. We used *p*-Ps annihilation gamma rays because the intrinsic lifetime of the *p*-Ps (0.125 ns) is much shorter than the slowing down process. Since the DB of the *p*-Ps is narrower than those from other annihilation processes, we could extract the *p*-Ps component from the energy spectrum of annihilation photons.

The Ps slowed down from 3.7 eV to 0.6 eV for 0.8 ns. Comparing with a kinetic model of Ps slowing down, we determined momentum transfer cross sections (σ_m) below 3.7 eV. We obtained σ_m in the Ps kinetic energy ranged from 2.0 to 3.8 eV for the first time. Present result agreed with the experimental result (below 2 eV) and calculation. In Ar gas at a pressure of 7.5 MPa, Ps formed with the kinetic energy of 3.7 eV and thermalized for 3 ns.

P63 Profile measurements of bremsstrahlung γ-ray for radioactive isotope production via photonuclear reaction at Research Center for Electron Photon Science, Tohoku University

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Nuclear chemistry and radioactive analysis is one of the major research fields in Research Center for Electron Photon Science, Tohoku University. Radioactive isotopes have been produced by photonuclear reaction with using electron linac. Bremsstrahlung γ-ray is created by the electron beam with the averaged current more than 100 μA injected to platinum or tungsten target. To answer various experimental requirements, the profiles of Bremsstrahlung γ-rays were studied with the current experimental configuration. It was obtained at the target position by irradiating Nickel films with 10 μm in thickness. An energy range of measured γ-rays was from 10 to 35 MeV within which photonuclear cross section of Nickel exists. ^{57}Ni Intensity and distribution were measured by a Ge Semiconductor detector and Phosphor imaging plates.

Measured FWHM of Bremsstrahlung γ-ray is 12 mm at the target position by injecting electron beam with FWHM width 7 mm to two 1 mm thickness Tungsten converters. The irradiation efficiency is approximately 7 % for the current sample size of a radioactive isotope production. By Geant4 simulation with (γ, n) cross section, the radioactivity of ^{57}Ni after the irradiation is estimated as 1 MBq integrating all irradiated area.

P64 Primary processes of radiolysis of sub- and super-critical water

Radiolytic products of coolant material under strong radiation field in water-cooled reactors are known to give undesirable effects on nuclear structural materials. Understanding of the fundamental processes will be of great importance for various application fields in water chemistry. Ionization and excitation of water molecules by ionizing radiations initiate very fast physical and chemical processes. Because of so high reactivity (short lifetime), it was difficult to observe experimentally the temporal behaviors (spatially inhomogeneous reactions, called spur diffusion reactions). In this work, the fundamental processes of the radiolysis of water at high temperature and pressure conditions (HTHP) were investigated by a newly developed picosecond time-resolved pulse radiolysis system, and also by numerical analyses. The results indicated that the hydrated electron (e-aq) in the spur reaction process mainly reacts with OH at room temperature, while that with H3O+ becomes also competitive in subcritical water. Taking the cumulative yield variations (ΔG molec./100eV) into account, it is suggested that historically defined primary G-value of the hydrated electron in subcritical water (G ~ 3.6 molec./100eV) will be appropriately reexamined to the lower value below 2.7 in neutral pH condition, while it is rather close to it in basic condition.

2015 日本放射化学会年会・第 59 回放射化学討論会
研究発表要旨集

編集・発行：
2015 日本放射化学会年会・第 59 回放射化学討論会
実行委員会（代表：関根　勉）
事務局：
東北大学大学院理学研究科化学専攻 放射化学研究室内
木野康志、岡　壽崇
（〒980-8578 仙台市青葉区荒巻字青葉6-3）
発行日：2015年9月12日（オンライン）