

English Abstracts

1S01 The forefront of diagnostic imaging and therapies in nuclear medicine

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Diagnostic nuclear medicine is as useful as the other imaging modalities as CT, MRI and ultrasonography. Particularly, its utilization has been expanded after the appearance of PET/CT and SPECT/CT. Although there are many diagnostic nuclear medicine procedures for all the organs, the diagnostic imaging for tumors, brain and heart of actual clinical cases will be presented in this lecture.

Radionuclide therapies include ^{131}I internal therapy for thyroid cancer and hyperthyroidism, ^{89}Sr (Metastron) internal therapy for pain from bone metastases, ^{90}Y (Zevalin) internal therapy for B-cell non-Hodgkin lymphoma, and brachytherapy using ^{125}I -seeds (OncoSeed) for prostate cancer. Besides, there are ^{131}I -MIBG internal therapy, boron neutron capture therapy (BNCT) for brain tumors and malignant melanoma, ^{177}Lu -octreotate therapy for neuroendocrine tumors, therapy with ^{153}Sm -EDTMP (Quadramet) or ^{223}Ra (Xofigo) for pain from metastatic bone tumors etc. These will be presented in this lecture.

2S01 Radiochemistry at J-PARC

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J-PARC (Japan Proton Accelerator Research Complex) is a high intensity proton beam facility, consisting of three proton accelerators and laboratories using the proton beam where secondary particle beams of neutron, muon, K-meson, neutron are available. MLF (Materials and Life Science Experimental Facility) uses a 3 GeV proton beam to produce intense muon beams and spallation neutron beams. The muon

beam is mainly used for μSR in study of solid state physics as well as fundamental physics experiments in particle and atomic physics. A new ultra slow muon beam line has been constructed in order to open a new research area of μSR . The strong neutron beams are used for developing new materials and also for life-related materials. Overview of the present status of J-PARC will be presented in order to encourage young and motivated people to join to use the unique and useful facility.

1A01 Neutron-induced nuclear reactions for Cu and Nb target at 134 and 184 MeV

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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 > 100 MeV have scarcely been undertaken because of the difficulty in obtaining high energy monoenergetic neutrons. Our group has developed a method for determination of high energy monoenergetic neutron cross-sections 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 134 MeV and 184 MeV have been studied for Cu and Nb targets. Excitation functions of short-lived radionuclides were determined by gamma-ray spectrometry. We also measured cross-sections by proton-induced reactions with the same energy range and compared with each other. The cross-sections of neutron-rich nuclides by neutron-induced reaction were systematically larger than these by proton-induced reactions at the same projectile energy.

1A02 Study on hot fusion reaction of $^{248}\text{Cm}+^{48}\text{Ca}\rightarrow^{296}\text{Lv}^*$ using GARIS

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We searched for a superheavy element (SHE) produced by hot fusion reaction of $^{248}\text{Cm}+^{48}\text{Ca}\rightarrow^{296}\text{Lv}^*$ using a gas-filled recoil

separator GARIS. This is the first study on production of the SHE with $Z\geq 113$ after we finished to study on the cold fusion reaction of $^{209}\text{Bi}(^{70}\text{Zn},n)^{278}113$ using GARIS. Although a trigger rate of 75 cps at focal plane in the $^{248}\text{Cm}+^{48}\text{Ca}$ reaction was relatively higher than a few cps in the $^{209}\text{Bi}+^{70}\text{Zn}$ reaction, we observed 5 decay chains by accumulating the beam dose of 4.3×10^{18} during a net irradiation time of 10 days. Decay characteristics of those events well agree with the precedent studies by both Dubna-Livermore collaboration and GSI SHIP collaboration. It is rather difficult to identify the nuclides of the products only from the present experimental study because we measured at only excitation energy. However, referring to the assignments of the precedent studies, we could state that two of the events are attributed to the decays of ^{293}Lv (3n evaporation channel), and three of them are attributed to the decays of ^{292}Lv (4n).

In this report, we will give these details including performance of background level and transmission of GARIS for hot fusion reaction.

1A03 Excitation functions for production of Nb and Ta isotopes in the $^{\text{nat}}\text{Zr}(d,x)$ and $^{\text{nat}}\text{Hf}(d,x)$ reactions

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The isotopes ^{95g}Nb ($T_{1/2} = 35$ d) and ^{179}Ta ($T_{1/2} = 1.82$ y) are useful radiotracers for the basic chemical studies of the element 105, Db. Previously, we investigated the production of these radiotracers by the activation of $^{\text{nat}}\text{Zr}$ and $^{\text{nat}}\text{Hf}$ with a 14-MeV proton beam supplied by the RIKEN AVF cyclotron. In this work, we

investigated the production of Nb and Ta isotopes in the deuteron-induced reactions on ^{nat}Zr and ^{nat}Hf .

A stacked-foil technique was used for the measurement of the excitation functions. The stacks of the thin foils of $^{nat}\text{Zr}/^{nat}\text{Ti}/^{nat}\text{Ta}$ and $^{nat}\text{Hf}/^{nat}\text{Ti}$ were irradiated for 30 min with 24-MeV deuterons supplied by the RIKEN AVF cyclotron. After the irradiation and proper cooling duration, γ - and X-rays of each foil were measured by Ge detectors.

The cross sections of $^{90g,91m,92m,95m,95g,96}\text{Nb}$, $^{95,97}\text{Zr}$, and $^{87m,87g,88}\text{Y}$ were measured in the $^{nat}\text{Zr}(d,x)$ reactions, while those of $^{175,176,178,179,180g}\text{Ta}$ and $^{175,179m2,180m,181}\text{Hf}$ were measured in the $^{nat}\text{Hf}(d,x)$ reactions. The cross sections of the $^{nat}\text{Hf}(d,x)^{179}\text{Ta}$ reaction were measured for the first time in this work, and the excitation function exhibits the maximum cross section of 489 ± 50 mb at 21.1 ± 0.4 MeV. For the most of the observed nuclides, only the partial agreements were found between the measured cross sections and the calculated ones by the TALYS-1.4 code, unlike our previous studies of the proton-induced reactions.

1A04 On the improvement of material properties and performance of nuclear targets

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Nuclear targets are frequently produced by means of the so-called molecular plating (MP) technique, which allows the quick electrodeposition of the element of interest from an organic medium with quantitative yields. Yet MP-produced targets are usually defective, characterized by surface cracks, and sometimes with poor structural rigidity. In order to improve the quality of the targets, constant current density MP experiments with lanthanides (i.e., mainly Nd and Sm) were carried out. Several plating parameters were varied and their effects on the properties of the produced deposits were investigated. It was found that by using very smooth deposition substrates and low volatility solvents smooth defect-free layers can be produced. The performance of defect-free ^{147}Sm layers used as α -particle sources for spectroscopy experiments proved to be superior to that of conventional targets as shown by 15% increases in relative detection efficiency. The optimized MP procedure was applied also for the preparation of homogeneous large-area (i.e., 42 cm^2) ^{242}Pu targets with minimum defectivity.

1A05 Investigation of p-process around mass number 140 (I) - $^{147}\text{Sm}(\gamma,p)$ reaction and its reverse process-

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The p-nuclei, which are not produced by neutron capture process, exist in the present solar system with low abundance. Nuclear reactions with a neutrino and charged particle

are considered to be dominant process for nucleosynthesis of the p-nuclei. We have attempted to establish abundance of the p-nuclei in mass number 130–150 using proton capture reactions. In stellar environment, nuclear reactions on excited nuclei should be considered. In the present work, the (p, γ) reaction cross section on excited state of ^{146}Pm was deduced from $^{147}\text{Sm}(\gamma,p)^{146}\text{Pm}$ reaction.

Samarium samples were irradiated with bremsstrahlung with end point energies of 10–50 MeV. Cross section of the $^{147}\text{Sm}(\gamma,p)^{146}\text{Pm}$ was determined from ^{146}Pm activity measured with a HPGe detector and bremsstrahlung spectrum estimated with Geant4 code. Cross sections of the reverse process on excited ^{146}Pm nuclei, namely $^{146}\text{Pm}(p,\gamma)^{147}\text{Sm}$ reaction, were estimated with a detailed balance theorem. The cross sections on the excited nucleus estimated from the (γ,p) reaction agreed with those estimated by a theoretical calculation using the TALYS code at energies around 6 MeV for proton energy. We concluded that large correction was not required due to deviation of cross sections on excited nuclei from those on the ground state.

1A06 Investigation of p-process around mass number 140 (II) -(p, γ) cross section measurements on proton rich nuclei-

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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. There are 35 stable p-nuclei heavier than iron, located between ^{74}Se and ^{196}Hg . Up to now, some nucleosynthesis processes of p-nuclei are proposed, for example (γ, n), (p, γ), and (ν_e, e^-) reactions and so on, but these reactions failed to describe the present isotopic ratios of p-nuclei in the solar system, yet. One of improvements to make is renewal the cross section data which are necessary to perform model calculations of the p-process.

For that purpose, we measured (p, γ) and (p, n) reaction cross sections on several p-nuclei around $A = 140$. Targets of ^{136}Ce , ^{138}Ce , and ^{144}Sm were irradiated with low-energy proton beam at Research Center for Nuclear Physics, Osaka University. The experimental cross section of $^{144}\text{Sm}(p, \gamma)$ was determined to be a value 10 times larger than a theoretical calculation using NON-SMOKER code at the lowest energy. The deviation may make a notable difference of isotopic abundances of the relevant nuclei in the model calculation. We will show how the discrepancy with the calculation affects an evolution of isotopic abundance.

1A07 Investigation of p-process around mass number 140 (III) -model calculation of isotopic abundance based on proton capture reaction-

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The p-nuclei which are unable to be produced by neutron capture process exist in the present solar system with low abundance. Nuclear reactions with neutrinos or charged particles are considered to be dominant processes for nucleosynthesis of the p-nuclei. We have

attempted to reproduce the current abundance of the p-nuclei in mass number 130 – 150 using proton capture reactions. In the present work, we performed a model simulation for the birth of the p-nuclei present in the solar system.

First, we computed by using the reaction rate isotopic abundances of Ba, Nd and Sm produced in s-process, which were not affected by the r-process. Then, we computed time variations of abundances of the p-nuclei produced at $(0.5 - 2.0) \times 10^9$ K for temperature and in 4×10^8 n/cm³ for neutron density assuming $10^2 - 10^6$ g/cm³ for proton density. The NON-SMOKER code was used for the reaction rate in the simulation.

A long-term nucleosynthesis shows a discrepant trend between ¹³⁶Ce/¹³⁸Ce ratio and ^{136,138}Ce/¹³⁴Ba ratio in any temperature and proton density. In contrast, all the abundance ratios of p-nuclei estimated in the work, ¹³⁶Ce/¹³⁸Ce, ^{136,138}Ce/¹³⁴Ba, ¹³⁸La/¹³⁴Ba, ¹⁴⁴Sm/¹⁴²Nd, are explained well with a short-term nucleosynthesis at $(1.2 - 1.5) \times 10^9$ K for temperature.

1A08 Coprecipitation behavior of element 104, Rf, with Sm hydroxide

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For coprecipitation experiment on element

104, Rf, we previously studied coprecipitation behaviors of its homologous elements, Zr and Hf, and pseudo homologue, Th. In addition, we developed an apparatus for rapid and repetitive preparation of precipitated samples. In this work, we performed online coprecipitation experiments using the apparatus for the ⁸⁵Zr and ¹⁶⁹Hf isotopes, which were produced with the AVF cyclotron at RIKEN and transported continuously from the nuclear reaction chamber to the chemistry laboratory with a He/KCl gas-jet system. Based on these results, the experimental conditions for Rf were determined. Then, we produced ²⁶¹Rf in the ²⁴⁸Cm(¹⁸O, 5n)²⁶¹Rf reaction and investigated its coprecipitation behavior with Sm hydroxide. Coprecipitation yields of Rf were determined as relative values to standard samples. Coprecipitation behavior of Rf was not similar to those of Zr and Hf but to that of Th.

1A09 Development of a rapid solvent extraction apparatus for rutherfordium

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So far, mainly solid-liquid extraction experiments have been performed in solution chemistry on superheavy elements. It is difficult to apply solvent extraction to short-lived superheavy elements because rapid equilibration and phase separation are needed to determine the distribution ratios (*D*) of superheavy elements. In this study, we developed a rapid extraction apparatus named “Flow Injection Extractor (FIE)” and phase separator to accomplish the solvent extraction and phase separation within

the lifetime of ^{261}Rf (68 s).

FIE is a flow-type extraction apparatus and consists of a Teflon tube filled with Teflon beads. Solvent extraction of ^{88}Zr and ^{175}Hf (homologues of Rf) was performed using 7.8-11.3 M hydrochloric acid as aqueous phase and 0.01 M Aliquat 336 benzene or carbon tetrachloride solution as organic phase. Two phases passed through FIE and then the mixture was collected in a test tube. The two phases were separately pipetted in tubes after centrifugation and were subjected to gamma ray measurement. The D values were determined from the gamma ray counts. The D values determined with FIE were consistent with those in equilibrium. This means that extraction equilibrium was achieved within 1 min by using FIE. In addition, we developed a flow-type phase separator using a PTFE filter. We found that separation with the purity of each phase of higher than 95% was achieved with a 1-step separator.

1A10 Extraction behavior of Nb and Ta with Aliquat 336 in HF solution

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We are planning to investigate the chemical properties of Db. In this experiment, ion-pair extraction of Nb and Ta, which are the lighter homologs of Db, with quaternary ammonium salt (Aliquat 336) from HF solutions is carried out to study complex formations of these elements with fluoride ions.

Radiotracers, ^{95g}Nb and ^{179}Ta , were produced

by proton bombardments on $^{\text{nat}}\text{Zr}$ and $^{\text{nat}}\text{Hf}$ metallic foil targets, using the AVF cyclotron at RIKEN and Tohoku University. These radiotracers were chemically isolated by an ion-exchange method. The tracers were dissolved in 600 μL of 0.01-27 M HF solution and then mixed with the same volume of 10^{-4} M Aliquat 336 in 1,2-dichloroethane solutions in a tube. After shaking the solutions for 5 min, and centrifuging for 5 min, the two phases were separately pipetted into sample tubes. The radioactivities of the two samples were measured with a Ge detector. Distribution ratios (D) were obtained from the ratio of the radioactivities in the two phases.

As the result, while the maximum D value of Ta was obtained in 0.27 M HF, that of Nb decreased gradually with increasing HF concentration from 0.01 M to 27 M. This suggests that there were clear differences between the chemical form species of Nb and Ta.

1A11 On-line reversed-phase chromatography of group-5 elements, Nb and Ta, for the chemical experiment of dubnium (Db)

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We have studied the solvent extraction and reversed-phase extraction behaviors of Nb and Ta with tributylphosphate (TBP) from hydrofluoric acid solutions for the chemical experiment of dubnium (Db). Since these results

showed that the extraction behavior of Ta is quite different from that of Nb, it is interesting to carry out chemical experiment of Db in this experimental condition. In this study, we performed on-line reversed-phase chromatography of Nb and Ta with Automated Rapid Chemistry Apparatus (ARCA), using short-lived isotopes of $^{90\text{g}}\text{Nb}$ and ^{178}Ta produced at the RIKEN AVF cyclotron. The reaction products transported by a He/KCl gas-jet system were dissolved in 1 M HF at a flow rate of 1 mL/min. The solution was fed into the extraction column of ARCA filled with 62-wt. % TBP resin, and the effluents were collected in plastic tubes. These samples were then subjected to γ -ray spectrometry using a Ge detector. Niobium was not adsorbed on the TBP resin, while Ta was retained. The present results are consistent with those obtained in our prior batch experiments. We will carry out the on-line experiment of Db.

1A12 Adsorption behavior of a superheavy element Rf by a TTA-reversed-phase chromatography method

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We study reversed-phase-chromatographic behavior of a superheavy element, Rutherfordium (Rf) with TTA as an extractant to clarify chemical properties of its cationic fluoride-complex. A nuclide of ^{261}Rf with a half-life of 68 s was produced in the $^{248}\text{Cm}(^{18}\text{O}, 5n)^{261}\text{Rf}$ reaction at the RIKEN K70 AVF

cyclotron. Nuclear reaction products were rapidly transported with a KCl/He gas-jet system to the chemistry laboratory, and then TTA-chromatographic behavior of Rf in acidic HF/HNO₃ solution was investigated with an on-line automated chemistry-apparatus (ARCA) and alpha-particle measurement system (RIDER). As a preliminary result, 17 counts (7.82-8.38 MeV) and 10 counts (7.82-8.38 MeV) of ^{261}Rf and its daughter ^{257}No were observed in the 1st and 2nd effluent samples, respectively, under an elution condition of 6×10^{-4} M HF/0.01 M HNO₃. This demonstrates that a cationic complex of Rf is formed in the solution and is extracted into the TTA-resin under the present conditions. In the symposium, adsorption probability of Rf onto the resin will be discussed in detail by comparing with those of Zr and Hf.

1A13 Study of mass asymmetric fission of the ^{193}Ir

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In the nuclear fission, mass distribution of fission fragments changes with the fissioning nucleus systematically, that is, an asymmetric mass distribution appears in many actinide nuclides while lighter nuclides lead to a symmetric fission. The cause of the asymmetric is that one fragment can easily become the nucleus in the neighborhood of ^{132}Sn which is the doubly closed shell nuclide. On the other

hand, in the case of a fission of a lighter nucleus, the nuclear fission such as liquid drop comes dominant.

It is expected that the mass distribution for the fission of ^{180}Hg is symmetric because ^{90}Zr has the neutron magic ($N = 50$) and the proton semimagic ($Z = 40$). In the beta-delayed fission of ^{180}Tl , however, an asymmetric mass distribution was observed by Andreev et al. This phenomenon is a new type of asymmetric fission and may not be caused by strong shell effects of the fragment, as observed in the actinide region.

Much theoretical investigation has been performed since this discovery. According to one result, existence of a unique asymmetry fission mode was predicted in the region of neutron-rich nuclei around iridium and tungsten. We took note of this prediction and carried out the experiments to find out a new asymmetric fission mode. In the presentation, we would like to talk about the results of the experiment in the reaction of $^7\text{Li} + \text{W}$.

1A14 Variation measurements of decay constants of Tc-99m as a chemical tracer

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The decay constants of some nuclides which decay via the internal conversion process are changed with changing environmental factors such as its chemical states. Example of this type of nuclides is metastable state of Tc-99 (Tc-99m). There is possibility that the variation measurement of the decay constants with radiation detection is able to use a chemical tracer. In this study, an apparatus for precise measurement of the decay constants was developed at first. The decay constants of

Tc-99m were precisely measured with various chemical state to investigate the effects of chemical states on the decay constants of nuclides. As a results, the difference of the decay constants between the two chemical forms, pertechnetate(VII) and hexachlorotechnetate(IV), is obtained on the order of magnitude of 0.1%. This indicates that the variation of the decay constants of Tc-99m is affected by the difference of the ligands rather than that of the oxidation state of Tc.

1A15 Performance test of a gas-filled recoil separator GARIS-II for asymmetric fusion reactions

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We have developed a new gas-filled recoil ion separator GARIS-II to study on asymmetric actinide-target based fusion reactions (hot fusion). It will be used as a powerful tool for a new superheavy element (SHE) search with $Z \geq 119$, SHE chemistry, studies on nuclear reaction and nuclear structure of SHE nuclide, and a direct mass measurement of SHE nuclide. The first quadrupole magnet of GARIS-II for vertical focusing is important role to increase a solid angle of the separator. Therefore, the separator is expected to have a high transmission for a hot fusion. We have already tested the separator using a standard α -source of ^{241}Am and

⁴⁰Ar-induced fusion reactions to evaluate the separation capability of GARIS-II from background particle and its transmission. These reaction products were properly collected onto the focal plane detection system with high efficiency under extremely low background condition. As a next step, on-line operating test of GARIS-II was performed using more asymmetric ²²Ne-induced fusion reactions of ¹⁹⁷Au and ²⁰⁸Pb. In this work, we tried to use He-H₂ mixture as a filled gas of the separator for the first time. In this report, we will talk about status of GARIS-II R&D and give the results obtained from studies on ²²Ne-induced fusion reaction in detail.

1A16 Measurement of the first ionization potential of nobelium (No, Z=102)

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The first ionization potentials (IPs) of heavy elements with atomic number $Z > 100$ have not been measured due to the short half-life and the low production rate of the isotopes. In order to determine the IP of the heavy elements, we have developed a novel measurement method based on a surface ionization technique by using a surface ionization ion source coupled to a He/CdI₂ gas-jet transport system for an Isotope Separator On-Line (ISOL) at the JAEA tandem accelerator facility. In this work, we have determined IP value of No by using the method. 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 present system, we measured ionization efficiencies of short-lived isotopes, ⁸⁰Rb, ¹⁵⁷Er, ¹⁶²Tm, ¹⁶⁸Lu, ^{142,143}Eu, ^{143m}Sm, ¹⁴⁸Tb, ^{153,154}Ho, ¹⁶⁵Yb, and ⁴⁹Cr produced in the reaction of ^{nat}Ge+¹¹B, ¹⁵¹Eu/¹⁵⁶Gd/¹⁶²Dy+¹¹B, ¹³⁶Ce/¹⁴¹Pr/¹⁵⁹Tb/¹⁴¹Nd/¹⁴⁷Sm+¹¹B, and ⁴⁵Sc+¹¹B. Ionization efficiency of ²⁵⁷No produced in the ²⁴⁸Cm(¹³C, 4n) reaction was also measured. Measured ionization efficiency of ²⁵⁷No was 0.8 %, which yields IP value of No to be 6.6 eV. This value is in a good agreement with the value which has been evaluated by extrapolation from those of the lighter actinide elements, 6.65 eV.

1A17 Electrolytic reduction of Mo and W as lighter homologues of seaborgium (Sg)

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We have been studying extraction and reduction behavior of Mo and W as preparatory experiments of a reduction of their heavier homolog, seaborgium (Sg). In this study, we searched for the reduction conditions from 6+ to 4+ ions of Mo and W and the separation ones

between the 4+ and 6+ ions which are required for our future on-line experiment of Sg. In experiments, we carried out electrolytic reduction of ^{93m}Mo and ^{176}W radiotracers produced at the JAEA tandem accelerator in such as $\text{H}_2\text{SO}_4/\text{HClO}_4$ solution. This was then followed by solvent extraction with TOA to identify the reduction of these elements on a tracer scale. Results showed that Mo is reduced in $\text{H}_2\text{SO}_4/\text{HClO}_4$ at around < -0.4 V while W is not reduced between 0.4 V and -1.4 V. In the presentation, details of the reduction and extraction of Mo and W are reported.

1B01 Evaluation of Redox Conditions at Dredged Trenches in Tokyo Bay by Mössbauer Spectroscopy and Neutron Activation Analysis

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In this study, the redox conditions at two dredged trenches in Tokyo Bay were evaluated for severe hypoxia, which has been observed every summer. The concentrations of U, Th, Ce, Fe, and Mn in Tokyo Bay sediments were measured by instrumental neutron activation analysis (INAA). The chemical states of iron were analyzed by ^{57}Fe Mössbauer spectroscopy.

In the Mössbauer spectra of the sediments, pyrite was detected in middle and lower layers of a dredged trench and in all layers below the surface of the non-dredged seabed. Therefore, both sites can be considered reductive, thereby fostering pyrite generation. The sedimentary environment was concluded to be more reductive in the seabed than in the trenches

because the pyrite ratios were higher in the seabed.

The values of Th/U and Ce/U ratios obtained from INAA were larger in the upper layers at both sites. Therefore, we concluded that the sedimentary environment of the upper layers is more oxidative than that of the lower layers. Since the U(VI)/U(IV) ratio was lower in the seabed, it is thought that the sedimentary environment was more reductive there, which is consistent with the results of the Mössbauer measurements.

Results obtained from Mössbauer spectroscopy of pyrite content proved comparable to those obtained by INAA. However, using both in conjunction allows for increased precision in estimation of redox potential in sediment.

1B02 Difference of spin state in anion-mixed assembled crystals bridged by 1,2-bis(4-pyridyl)ethane

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New anion-mixed complexes using KNCS and NaNCBH_3 having iron as metal and 1,2-bis(4-pyridyl)ethane as bridging ligand were synthesized. These complexes have a trend to have more NCS than the fraction as synthesized. Powder X-ray diffraction pattern shows (2 0 -2) and (1 1 -2) diffraction, shifting to higher degree with increasing fraction of NCS. All the Mössbauer spectra basically consist of two doublets, outer and inner doublets. Q.S. value of outer doublet changes depending on the anion fraction. The values are relatively similar to the original pure anion complexes. On the other hand, Q.S. value of inner doublet is independent of anion fraction, but the origin is unknown.

There is a possibility of mixed complex having one NCS and one NCBH_3 as counter anion, although Q.S. of the iron with one NCS and one NCBH_3 is expected to have larger value. While the anion-mixed complexes bridged by 1,3-bis(4-pyridyl)propane did not show the spectrum having one NCS and one NCBH_3 . The difference between 1,2-bis(4-pyridyl)ethane complex and 1,3-bis(4-pyridyl)propane is also unknown. Mixed complexes including 1,4-dichlorobenzene were also obtained. Mössbauer spectra revealed more low-spin state compared with the sum of low-spin state in each pure complex.

1B03 Synthesis and Mössbauer Spectroscopic Observation of Valence-trapping and Detrapping of two Polymorphs of Mixed-valence Trinuclear Iron Pentafluorobenzoate complex

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We have studied behaviors of iron-valence trapping and detrapping phenomenon in mixed-valence trinuclear iron carboxylate complexes. In our previous Mössbauer work, it was revealed that three iron ions in $\text{Fe}^{3+}\text{Fe}^{3+}\text{Fe}^{2+}\text{O}(\text{C}_6\text{F}_5\text{COO})_6(\text{C}_5\text{H}_5\text{N})_3\cdot\text{CH}_2\text{Cl}_2$ are in the valence-trapped state in lower temperatures, while in the valence-detrapped state in higher temperatures, the transition temperature being around 80 K. A single-crystal X-ray diffraction analysis at 123 K revealed that this mixed-valence complex crystallized in a hexagonal system and three iron atoms were equivalent in consistent to the Mössbauer

observation. Very recently, new-type crystals having the same chemical formula $\text{Fe}_3\text{O}(\text{C}_6\text{F}_5\text{COO})_6(\text{C}_5\text{H}_5\text{N})_3\cdot\text{CH}_2\text{Cl}_2$ was obtained by a very slow recrystallization from a dichloromethane solution. A powder X-ray diffraction showed a different diffraction pattern to that of the hexagonal crystals, suggesting a polymorph. The new polymorph could be assigned to orthorhombic from the XRD pattern. Temperature-dependent Mössbauer spectra of the new-type complex showed that the iron valences were in the trapped state in a wide temperature range from 78 to 300 K, in remarkable contrast to the hexagonal one.

1B04 Determination of Formation Energy of Oxygen Vacancies in Zinc Oxide Doped with Al and In Impurities

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The time-differential perturbed angular correlation method was applied to a study of local fields at the $^{111}\text{In}(\rightarrow^{111}\text{Cd})$ probe in Al-doped ZnO. 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. Furthermore, it is suggested from another TDPAC measurement for 100 ppm Al-doped ZnO annealed in Ar gas that the local association of Al and ^{111}In becomes unstable in anaerobic atmosphere due to the formation of oxygen vacancies in the sample. In order to provide

further insight into the dissociation mechanism, in the present work, we analyzed TDPAC spectra for 100 ppm Al-doped ZnO prepared on various heat-treatment conditions, and found that the relevant atmosphere dependence of the impurity behaviors provides a helpful clue to the dissociation mechanism triggered by the formation of oxygen vacancies.

1B05 Dynamics of ^{111}Ag in a superionic conductor AgI

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Silver iodide (AgI) offers superionic conductivity expected for future technological applications. This conducting phenomenon, however, emerges only in its high-temperature α phase (≥ 420 K), which has long been a barrier to practical applications of this compound. Recently, an epoch-making technique overcame this situation: AgI powder coated by poly-N-vinyl-2-pyrrolidone (PVP) can partially preserve the conducting α phase even at room temperature. For detailed understanding of the low temperature conductivity, in the present work, local dynamics of Ag^+ ions was investigated by means of the time-differential perturbed angular correlation (TDPAC) technique using the $^{111}\text{Ag}(\rightarrow^{111}\text{Cd})$ probe.

The probe was separated by an anion-exchange chromatography from Pd matrix irradiated with thermal neutrons in KUR. The isolated ^{111}Ag was then incorporated in AgI powder when it was synthesized by precipitation with PVP powder. TDPAC measurements were

then performed at various temperatures down to room temperature.

All the TDPAC spectra show exponential relaxation reflecting dynamic perturbation ascribable to hopping motion of Ag^+ ions. It is considered that the PVP-coating has made it possible for the α phase to survive even at room temperature. We here report successful atomic-level observation of room-temperature dynamic motion of Ag^+ ions, and discuss the activation energy of the local dynamics.

1B06 Medical Radioisotope Production with Accelerator Neutrons by 40 MeV 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 ^{99}Mo ($T_{1/2} = 66$ h), ^{90}Y ($T_{1/2} = 64$ h), ^{67}Cu ($T_{1/2} = 61.8$ h), and ^{64}Cu ($T_{1/2} = 12.7$ h). Enriched ^{100}Mo , ^{90}Zr , ^{68}Zn and ^{64}Zn oxide samples were irradiated with neutrons, which were obtained by the $^{12}\text{C}(d,n)$ and $\text{Be}(d,n)$ using 40 MeV deuterons provided from the TIARA cyclotron. ^{99}Mo , ^{90}Y , ^{67}Cu , and ^{64}Cu were successfully produced via the (n, x) reactions, and we clearly observed the γ -rays. Particularly, yields of impurity radionuclides were much smaller than that of ^{99}Mo and ^{67}Cu , and therefore radioactive waste produced during chemical processing would be reduced compared with other proposed reaction systems. The present results demonstrate that the radioisotopes, ^{99}Mo , ^{90}Y , ^{67}Cu , and ^{64}Cu , can be produced by using fast neutrons, and strongly suggest that the

reaction system is one of the most promising routes to produce high quality medical radioisotope.

1B07 Production and utilization of astatine and iodine radioisotopes using ^7Li ion beams

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In general, an α -emitter ^{211}At which is a prospective candidate for utilization in targeted alpha radiotherapy is produced through the $^{209}\text{Bi}(^4\text{He},2n)^{211}\text{At}$ reaction. In contrast, our project is focusing on the production in the $^{209}\text{Bi}(^7\text{Li},5n)^{211}\text{Rn}$ reaction. This enables us to supply ^{211}At in a $^{211}\text{Rn}/^{211}\text{At}$ generator system. The daughter ^{211}At (7.2 h half-life) is extracted from the parent ^{211}Rn (14h), expanding time-frame for transportation and use of ^{211}At .

To use astatine and iodine radioisotopes in our project, the excitation functions of $^{209}\text{Bi}(^7\text{Li},xn)^{216-x}\text{Rn}$, $^{\text{nat}}\text{Pb}(^7\text{Li},xn)^{\text{nat-x}}\text{At}$ and $^{\text{nat}}\text{Sn}(^7\text{Li},xn)^{\text{nat-x}}\text{I}$ reactions have been measured. In addition to that, separation techniques have been developed. We report not only on the production and separation of astatine and iodine radioisotopes in the reactions but also on the utilization of those radioisotopes.

1B08 Production of ^{67}Cu for Cancer Therapy Using Fast Neutrons from $^{\text{nat}}\text{C}(d,n)$ Reaction

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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 for clinical researches has been limited due to the difficulty in obtaining sufficient quantities. In this study, we have investigated the production of ^{67}Cu by the $^{68}\text{Zn}(n,x)^{67}\text{Cu}$ reaction ($x=n'p, d$) using fast neutrons from $^{\text{nat}}\text{C}(d,n)$ reaction. The highly purified ^{67}Cu was obtained by the two columns separation method (chelate resin chelex-100 and anion exchange resin AG1-X8). Furthermore, the labelling of DOTA and TETA which are useful bifunctional ligands for the labelling monoclonal antibodies was succeeded in more than 97% yield.

1B09 Preparation of ^{191}Pt for cancer treatment

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The platinum-based anticancer agent like cisplatin is one of the most important carcinostatic, because of its broad spectrum anticancer activity. However, it is difficult to

produce RI-tracer of Pt, and the biodistribution of these drugs are still unknown. Then we aim to make the establishment of preparation of ^{191}Pt and visualize the uptakes of platinum-based anticancer drug. In this work, we investigate the production of ^{191}Pt via $^{nat}\text{Pt}(p, xn)$ reaction with 46 - 60 MeV proton beam from AVF cyclotron at Research Center for Nuclear Physics. Furthermore, several method to purify ^{191}Pt with liquid-liquid extraction was tested, and we obtained high radio specific activity ^{191}Pt which contained little radioactive impurities. Additionally, we synthesized ^{191}Pt -cisplatin, and observed the biodistribution of it in mice at the 1 and 24 h time point (each $n = 4$). This work indicates that our preparation method of ^{191}Pt is available for medical use.

1B10 Isolation of ^{211}At from an irradiated Bi target using a wet chemistry approach

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^{211}At (Half-life: 7.21 h) is one of the promising radionuclides for targeted alpha therapy. Dry distillation has been widely used to isolate ^{211}At from a Bi target. Poor reproducibility of recovery yield in the dry distillation led us to employ a wet chemistry approach. High and stable recovery yield ($\sim 78 \pm 11\%$) was accomplished by the approach. However, the use of diisopropylether (DIPE) in the isolation process has made it difficult to automate the process due to miscibility. Therefore, we have investigated column methods to simplify the isolation of ^{211}At . Anion exchange resin (AG1x8), cation exchange resin (AG MP-50), and polyethylene glycol (PEG) coated resin were used. The capture efficiency of

^{211}At on AG 1x8 was high when loading ^{211}At with 8M HCl (99%), but decreased when using boric buffer. Low capture efficiency was observed when AG MP-50 was used. High capture efficiencies (96-99%) were obtained with PEG-coated resin. Low recovery efficiency ($\sim 6\%$) from AG 1x8 was obtained using strong base solution. On the other hand, ^{211}At was recovered from PEG-coated resin in good efficiency (60-79%) when using NH_4OH solution as eluent. These studies demonstrated that PEG-coated resin could be used to isolate ^{211}At from a Bi target.

1B11 Evaluation of radon trapping and daughter recovery in organic solvent based system

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Radon-211(^{211}Rn), a noble gas radionuclide is a promising for supply an alpha emitting radionuclide astatine-211(^{211}At), which has been used for targeted alpha therapy in nuclear medicine, as a $^{211}\text{Rn}/^{211}\text{At}$ generator system. To make a suitable design of generator system, we choose wet chemistry and solvent extraction methods to collect ^{211}Rn and retain it in an appropriate solvent to ingrowth of ^{211}At . In this study, we evaluate trapping yield of ^{211}Rn in several organic solvents (such as DIPE, MIBK, toluene, hexane, decane, dodecane), and also evaluated back extraction yield of ^{211}At in methanol. The trapping yields of ^{211}Rn in

organic solvents were ca 70-90% and there seems no correlation in each selected solvent. The back extraction yields of ^{211}At in methanol 15 hour after ingrowth in hexane, decane, dodecane were ca 20-30%.

1B12 Conversion of Stable Geometrical Isomers between Pentavalent and Tetravalent Uranium Complexes with Salophen-type Ligand

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Our group recently synthesized the eight-coordinated dodecahedral U(V) complex with salophen-type ligands (*N,N'*-(1,2-phenylene)bis(5-methyl-2-hydroxybenzylideneimine)) by the one-electron oxidation of the eight-coordinated square-antiprismatic U(IV) complex. In the present study, we investigated in detail about the structural transformation by oxidation of the uranium center.

A brown precipitate of the U(IV) complex was obtained by the reaction of UCl_4 with 2 eq. of salophen-type ligand. When the precipitate was recrystallized from THF, platelet (1a) and block (1b) crystals were obtained. The single-crystal X-ray structure analysis revealed that the uranium ion in 1a is a square-antiprismatic structure, and that in 1b is a dodecahedral geometry. These structures tautomerized in solution, which was studied by the change in redox waves of 1a and 1b. The equilibrium constant defined as $[1b]/[1a]$ was 0.082 in CH_2Cl_2 . Therefore, a square-antiprismatic structure was more stable in the U(IV) isomers.

When 1a was chemically oxidized in CH_2Cl_2 ,

the U(V) complex was obtained. In the case of U(V), two geometrical isomers, a square-antiprismatic (2a) and a dodecahedral (2b) structures, tautomerized. The equilibrium constant defined as $[2b]/[2a]$ was 6.5 in CH_2Cl_2 . This indicates that the more stable structure of U(V) complex is the dodecahedral structure, unlike in the case of the isomorphous U(IV) complexes.

1B13 Complexation of ^{177}Lu with bifunctional chelators in the presence of competing metals

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As part of basic studies for the production of ^{177}Lu capable of labeling of ^{177}Lu -1,4,7,10-tetraazacyclododecan-*N,N',N''*,*N'''*-tetraacetic acid (DOTA)-antibody, ^{177}Lu complexation of DOTA and diethylenetriamine-*N,N,N',N'',N'''*-pentaacetic acid (DTPA) was investigated in the presence of competing metals: Mg, Cu, and Yb in addition to Ca, Fe, and Zn. The inhibition of ^{177}Lu complexation by the competing metals was in the order of Mg, Ca < Fe < Yb < Cu, Zn and Mg, Ca < Zn < Fe < Cu < Yb with DOTA and DTPA, respectively. Consequently, the elimination of Mg and Ca from the ^{177}Lu final solution produced was not found to be necessary, while the elimination of Cu, Fe, Zn and Yb from the ^{177}Lu final solution was found to be necessary.

2A01 Determination of antistatic properties of non-woven fabrics of protective clothing with measurement of α -particles from radioactive radon daughters

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Most tasks for restoring Fukushima Daiichi nuclear disaster force personnel to put protective cloths on. The radioactive contamination of those cloths increases radioactive dose of personnel and also makes difficulty in the waste disposal. As the contamination might mainly be caused from electrostatic adsorption, it is important to reduce the electrification of the protective cloths.

The Entira® film developed by Du Pont-Mitsui Polychemicals is not just anti-static but waterproof and breathable. By backing Entira film with suitable unwoven fabric, adequate material for protective cloths can be fabricated. However, as the contamination can be caused even by very weak electrification, unwoven fabric layer could destroy anti-static property of Entira film. Such weak electrification cannot be evaluated with the conventional method such as measuring the surface charge.

We tried to evaluate electrification by utilizing alpha measurement of Radon daughters. The materials are left in the Kyoto University Critical Assembly (KUCA) for a several weeks. Fine particles containing Radon daughters emitted from thick concrete walls of KUCA accumulate on the surfaces of those materials. The alpha emission was counted with the alpha survey meters. Large difference in alpha counting among different material was clearly observed. It can be considered that this method is effective for evaluating weak electrification.

2A02 Consideration of sub-criticality

monitoring method by measuring isotope ratio of FP rare gas –Focusing on collection of radioactive rare gas from the fuel debris–

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(NAIS Co., Inc.)

For Fukushima Daiichi nuclear power station, it is important to establish a sub-criticality monitoring method in order to prevent re-criticality of the fuel debris. The conventional technique is the neutron method, but it is difficult for the damaged reactor cores to detect neutrons due to high background and complicated structure. In such situations, the sub-criticality monitoring method by measuring FP rare gas is effective. As an example, the authors pay attention to the fission yield ratio of Kr-88 and Xe-135 which are produced by thermal neutron induced fission of U-235 and spontaneous fission of Cm-244. The ratio varies in approximately one digit: 1.86 for U-235 and 17.6 for Cm-244. Therefore it will be easy to estimate ratio of the number of U-235 neutron fission and the number of Cm-244 spontaneous fission by taking isotope correlation between Kr-88 and the Xe-135. In this presentation, collection of the radioactive rare gas are especially considered for the sub-criticality monitoring method by measuring isotope ratio of short lived rare gas nuclides, such as Kr-88 and Xe-135, from the fuel debris.

2A03 Isotopic composition of Uranium and Transuranium in the environment derived from Fukushima Dai-ichi Nuclear Power Plant accident

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Dust samples from the sides of roads (black

substances) have been collected together with litter and soil samples at more than 100 sites contaminated heavily in the 20-km exclusion zones around Fukushima Dai-ichi Nuclear Power Plant (Minamisoma City, and Namie, Futaba and Okuma Towns), in Iitate Village located from 25 to 45 km northwest of the plant and in southern areas from the plant. Isotopes of U, Pu, Am and Cm have been measured in the samples to evaluate their total releases into the environment from the FDNPP and to get the isotopic compositions among these nuclides. The results provided a coherent data set on $^{239+240}\text{Pu}$ inventories and isotopic composition among these uranium and transuranic nuclides. When these activity ratios were compared with those for fuel core inventories in the FDNPP accident estimated by a group at JAEA, except $^{239+240}\text{Pu}/^{137}\text{Cs}$ activity ratios, fairly good agreements were found, indicating that uranium and transuranic nuclides were released into the environment without their large fractionations. The obtained data may lead to more accurate information about the on-site situation (e.g., burn-up, conditions of fuel during the release phase, etc.), which would be difficult to get otherwise, and more detailed information on the dispersion and deposition processes of transuranic nuclides and the behavior of these nuclides in the environment.

2A04 Microscopic distribution of radioactive Cs in the phlogopite, using electron microscope autoradiography

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Radioactive material of a large amount has been released to the environment by the accident at the Fukushima Daiichi nuclear power plant. As a result, radioactive Cs adsorbed in the soil.

It is well known that radioactive Cs is easy to adsorption to clay with study of macro autoradiography (ARG). But, discussion is difficult for the relevance of adsorption and micro structure, because Imaging Plate method is low resolution. We tried to observe the microscopic distribution of radioactive Cs in phlogopite by Scanning Electron Microscope (SEM)-ARG. SEM-ARG is using the emulsion and observe by SEM.

Experimental results shows to observe the microscopic distribution of radioactive Cs on phlogopite at tens of micrometers. The form of the Ag that was exposed is different by development time and the radioactivity. Observed Ag grain size is 5 to 20 μm .

This method may be applicable to phlogopite as well as other materials. This method could be useful to elucidate the adsorption state of radionuclide.

2A05 Analysis of ^{93}Zr in the contaminated water generated at the Fukushima Daiichi Nuclear Power Station

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(Japan Atomic Energy Agency)

Recently, the separation method of Zr, Nb, and Mo from major elements of contaminated water, followed by mutual separation of Zr, Nb, and Mo using single solid phase extractant column, TEtra VAlent actinide (TEVA) resin column, has been developed to analyze ^{93}Zr . In this work, the separation method was applied to the analysis of ^{93}Zr in contaminated water generated at the Fukushima Daiichi Nuclear Power Station. Zirconium in the contaminated water sample and in ^{90}Zr spiked contaminated water sample was separated with the method and then measured by inductively coupled plasma mass spectrometry (ICP-MS). Recovery of Zr

determined by spiked ^{90}Zr was $100\pm 2\%$. Calibration curve of ^{93}Zr was induced from the slope and intercept of ^{92}Zr and ^{94}Zr . The count rate of mass number 93 of the Zr fraction was background level. Consequently, ^{93}Zr in the contaminated water was determined $< 0.1 \text{ ng/mL}$ (9 mBq/mL).

2A06 Behavior of Cesium-134, 137 and Iodine-129 in River

SUEKI, K.¹, SHIBAYAMA, N.², SATOU, Y.², SASA, K.¹, TAKAHASHI, T.³, MATSUNAKA, T.³, MATSUMURA, M.³, MATSUZAKI, H.⁴, MURAKAMI, M.⁵, YAMASHITA, R.⁶, SAHA, M.⁶, TAKADA, H.⁶, KOIBUCHI, Y.⁷, LAMXAY, S.⁷, O, H.⁷, MOURI, G.⁵, OKI, T.⁵

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Radioactive cesium and iodine which emitted from the Fukushima Daiichi nuclear power plant were detected from water in environment. In the metropolitan area, higher concentration of radioactive cesium was detected from sediment of the Ohori River, Chiba Prefecture. In this work, the long-term variations of the concentrations of the radioactive cesium in the suspended substance (SS) and dissolved matter (DM) were observed for the Ohori River, and the elucidation of the actual condition was tried. It was observed concentrations of ^{134}Cs , ^{137}Cs and ^{129}I in the SS and DM in the river. The concentration change of the average of the radioactive cesium contained in the SS was obtained by the SS sampler of the Ohori River installed at the mostly median center. In the

same place point, moreover, the river water sampled for biweekly and obtained the radioactive cesium concentration of the moment from May, 2012. It is thought which these time variations make it possible to evaluate the external factors over river such as rain. We obtained that $[\text{}^{137}\text{Cs}]_{\text{SS}}/[\text{}^{137}\text{Cs}]_{\text{DM}}$ ratio is 4.0×10^4 in the Ohori River. And it correlated well between the variation of the radioactive cesium concentration and its iodine-129 concentration in dissolved matter.

2A07 Autoradiography and $^{134}\text{Cs}/^{137}\text{Cs}$ ratios of SPM collected at automated SPM monitoring stations at the FDNPP accident

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Radioactivity ratios of $^{134}\text{Cs}/^{137}\text{Cs}$ of SPM collected SPM monitoring stations operated by local governments in eastern Japan on 11 to 23 Mar. 2011 were determined precisely and autoradiographs using an imaging plate were taken for the SPM samples to evaluate a distribution of radionuclides within SPM. Although most of SPM samples had 1.03 of $^{134}\text{Cs}/^{137}\text{Cs}$ ratio, some SPM had different ratios, 0.921 and 1.01. The ratio of 0.921 was found in only SPM collected at stations located on northern area to FDNPP on 12 Mar. 2011. Radiocesium having low $^{134}\text{Cs}/^{137}\text{Cs}$ ratio was expected to be released along with an accident at Fukushima Daiichi Unit 1 reactor on 12 Mar. and determined ratio, 0.921, in this work was close to evaluated ratio, 0.941, in the core of Unit 1 obtained by ORIGEN2 code by Nishihara et al. Ratio of 1.03 was observed in SPM collected on only 16 Mar. In autoradiography of SPM samples, fine particles with high

radioactivity were found in SPM collected on 15 Mar. at most of monitoring stations in eastern Japan. On the contrary, on 21 Mar., a few particles with high radioactivity were observed in each SPM samples.

2A08 Radioactive measurement of ^{140}Ba in air-dust samples collected in the early phase of FDNPP

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Barium-140 was one of radioactive nuclide discharged to the atmosphere by the Fukushima Dai-ichi nuclear power plant accident. The discharge behavior of ^{140}Ba was different from that of ^{137}Cs due to non-volatility property of barium. Also Barium-140 is the same chemical family with ^{90}Sr and have the similar behavior in fuels when the nuclear reactor normal operation. However, ^{140}Ba has more volatility than ^{90}Sr in the severe accident phase. Therefore, comparing the discharge amount of ^{137}Cs , ^{90}Sr and ^{140}Ba can bring us information about the accident circumstances.

We measured ^{137}Cs and ^{140}Ba activity in the air dust samples collected in the early phase of the FDNPP accident with germanium semiconductor detector. The sampling spots were Tsukuba, Hitachi and Mito cities, Ibaraki Prefecture and Kawasaki City, Kanagawa Prefecture. Strontium-90 was isolated from each sample with solid-phase extraction and ion-exchange separation and its activity was measured by Cherenkov light counting.

The $^{140}\text{Ba}/^{137}\text{Cs}$ activity ratio was 0.07 at 2011/03/15 and it rose about 0.15 in April. In the presentation, the discharge amount of ^{140}Ba will

be compared with that of ^{90}Sr by considering total inventory in the core.

2A09 Physical and Chemical Characteristics of Radio-Cs Emitted during the Early Stage of the Fukushima Nuclear Power Plant Accident

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We have investigated chemical and physical properties of the radioactive aerosol, from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident occurred in March 2011, by using imaging plate technique, scanning electron microscopy with energy dispersive X-ray spectroscopy, etc. (Adachi et al., 2013).

We have found more than 10 spherical Cs-bearing particles (Cs-ball) from HV filter samples collected during Mar. 14-15, 2011, when the first radioactive plume arrived from the FDNPP accident at the Meteorological Research Institute as well as Univ. Tsukuba, Japan. While the second plume gave no such Cs-ball. They have basically spherical morphology, characterized composition of Fe, Zn, Mn, O, etc. with an appreciable amount of Cs. They are a few microns in diameter with a few Bq of radio-Cs activity but as high specific activity as sub-terra Bq/g. They were found to be insoluble; even refractory to conc. nitric acid and soluble only to sodium hydroxide solution (Sueki et al., 2013). Also, the bulk extraction experiments showed that the Cs-ball could occupy major part in the first plume.

The synchrotron X-ray microbeam analyses by using SPring-8 facility suggested that the Cs-ball contains many possible fission products

in glass matrix (Abe et al., 2014). Therefore, they would persist for a long time in the environment, and we need to consider the impacts of the Cs-ball.

Aerosol model simulation gave the results that non-hygroscopic super-micron Cs-ball could be removed more by the dry deposition and below-cloud scavenging than conventional sulfate-hosted particles.

2A10 Classification of radioactive contamination according to $^{110m}\text{Ag}/^{137}\text{Cs}$ ratios in the northwest area of the Fukushima Daiichi NPP

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Activity ratio and isotopic ratio are one of essential information to determine a source of radionuclides. Activity ratio of $^{110m}\text{Ag}/^{137}\text{Cs}$ was varied at the northwestern area from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) by the MEXT survey. Three groups were recognized from the activity ratio at the northwestern area. The ratio of the one group, which located in the vicinity of the FDNPP corresponded to that of the unit 1 estimated with the ORIGEN 2 code. In contrast, the ratios of another two groups were lower than the estimation by one order because the area was contaminated after the nuclear core was cooled down. These groups would be affected from the unit 2 or 3. The present work indicates that three kinds of local contamination are observed from the activity ratio at the northwestern area from the FDNPP.

2A11 Muon transfer process for H_2+CO and

H_2+CO_2 mixtures in low pressure gaseous conditions

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A negative muon is one of the elementary particles that has charge and approximately two hundred times heavier mass than an electron. A negative muon can be captured by the Coulomb field of a nucleus, and then a muonic atom is formed. When the muon is captured by a hydrogen atom, muonic hydrogen atom which can be regarded as a pseudo neutron is formed. Muonic hydrogen atom can penetrate the electron cloud of heavier atom and gives the muon to the deeper atomic levels in the heavier nucleus. This process is called muon transfer. The muon capture processes by muon transfer process and direct capture process (negative muon is captured by heavier atom directly) are different each other, however, the difference on initial muon capture levels and muon capture probabilities by these processes still not been well examined. In this study, we performed muon irradiation experiments at J-PARC, Ibaraki, for low pressure CO , CO_2 , H_2+CO and H_2+CO_2 mixture gases, and measured muonic X-rays which are emitted after muonic atom formation. From these experiments, the initial muon capture levels and muon capture ratios were determined quantitatively for CO and CO_2 molecules by two muonic atom formation processes.

2A12 Muon transfer process from muonic hydrogen in gaseous benzene and cyclohexane

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The formation processes of muonic and pionic atoms are affected by the chemical environment of the muon- or pion-capturing atoms. Such a phenomenon is called chemical effect. Such a chemical effect was also reported in pion transfer process, but it has never been founded in muon transfer process. To examine the chemical effect on muon transfer process, we selected the gas mixture of C₆H₆ (0.2 kPa) + Ne (3.0 kPa) + H₂ (96.8 kPa) and C₆H₁₂ (0.2 kPa) + Ne (3.0 kPa) + H₂ (96.8 kPa) as the muon irradiation samples. The muon irradiation experiments were performed at the MLF/MUSE D1 beam line of the Japan Proton Accelerator Research Complex (J-PARC), Ibaraki, Japan. Muonic X-rays emitted after muon-capture for carbon or neon atoms were measured using high purity germanium detectors.

Muon transfer rates were determined from delayed component of the muonic X-rays. Delayed muonic X-ray intensity ratio of $\mu\text{C}/\mu\text{Ne}$ corresponds to the ratio of muon transfer rates for carbon atoms to that for neon atoms. The ratios of $\mu\text{C}/\mu\text{Ne}$ were 6.4 ± 0.7 in C₆H₆+Ne+H₂ sample and 5.7 ± 0.5 in C₆H₁₂+Ne+H₂ sample. We concluded that the significant difference in muon transfer rate for carbon atoms in two samples was not detected in this work.

2A13 Research of Fluctuating Valence-state in Mixed-valence Trinuclear Iron Carboxylate Complexes by Muon Spin Relaxation and Mössbauer Spectroscopy

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Both by muon spin relaxation (μSR) technique and ⁵⁷Fe Mössbauer spectroscopy, iron-valence fluctuation behaviors were probed in polycrystalline mixed-valence trinuclear iron carboxylate complexes. A Mössbauer study of Fe₃O(CH₃COO)₆(H₂O)₃·2H₂O indicated that the valence states of the three iron atoms were changed from the trapped one (Fe³⁺Fe³⁺Fe²⁺) at 78 K to the detrapped one (Fe^{2.7+}Fe^{2.7+}Fe^{2.7+}) with increase in temperature, and the transition temperature was estimated to be around 200 K. Above this temperature, valence fluctuation via electron-hopping was assumed to occur. μSR experiments were carried out at D1 muon-beam port in MLF of J-Parc and the muon spin relaxation rates (i.e., the decay constant of muon-spin polarization) were determined at 100, 170, 240, and 300 K under a transverse magnetic field of 20 G. A maximum value in the relaxation rates was observed at 170 K. A slow electronic fluctuation would occur around 170 K in Fe₃O(CH₃COO)₆(H₂O)₃·2H₂O, which was consistent with the above Mössbauer spectroscopic observation. The Mössbauer and μSR studies of Fe₃O(C₆F₅COO)₆(C₅H₅N)₃·CH₂Cl₂ were also discussed.

2B01 A study on pretreatment of tritium analysis in environmental samples using microwave heating method

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Industrial Science and Technology)

Tritium in environmental organic samples usually consists of free water tritium (FWT) and organically-bound tritium (OBT). The FWT is tritiated water existed as HTO form, and its behavior is similar to natural water. On the other hand, OBT is bound tritium to organic tissue. The OBT has a longer biological half-life and higher incorporation to organic materials than the FWT. The standard method of FWT and OBT analysis is a liquid scintillation counting method after freeze-drying and combustion of the sample as summarized in MEXT report. However, pretreatment / analytical method of FWT and OBT are complicated and take a long time over a week. In this paper, to simplify the analytical method for FWT and OBT, we report basic study of TG/DTA analysis of pine needle samples and the preliminary result of effective pretreatment for pine needle samples using microwave heating method.

2B02 Development of tritium detector system combined with proton conductor ceramic membrane

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The detection of low level tritium without radioactive waste such as liquid scintillation cocktail is one of the key issues for tritium management in tritium handling facilities. In order to detect a low level gaseous tritium, the enrichment of tritium by a membrane separator is an effective measure. The application of an electrochemical hydrogen pump using a proton conducting oxide that transports protons in oxides at high temperatures have been proposed as a membrane separator. We have reported the results of the system integration test under the conditions without oxygen gas. In this study, the

integration test under the practical condition using room air was conducted at Isotope Separation Laboratory as tritium handling facility in Nagoya University. The system mainly consists of proton conductor ceramic membrane, metal bellows pump, commercial proportional counter LB110 (Berthold Technology) and data logger LB5320. The estimated tritium concentration by the proposal system shows good agreement with the introduced tritium concentration. The result of this work is that the measurement of the tritium concentration via the proposed tritium monitor system was successfully demonstrated. Technically, the proposal system has reached a level at which it can be put into practical use.

2B03 Dynamics of tritium recovery systems using water bubbler and CuO catalyst

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Tritium is used as a tracer of hydrogen isotopes. However, the recovery of a small amount of tritium at radiation facility is usually unreasonable due to higher basic safety standards of tritium. To enhance the social acceptance, tritium should be recovered as much as possible without complex procedure. In this study, elucidation of reaction mechanism from tritium gas to tritiated water by CuO catalyst and evaluation of reaction rate constant were performed.

Tritium gas was recovered from a tritium ion irradiation device via a scroll pump. It was mixed with PR gas and introduced into the first proportional counter to measure over all tritium.

Thereafter, CuO was used to oxidize the gas including HT. The oxidized gas was completely recovered by a water bubbler. Finally, the gas was introduced into the second proportional counter to measure unreacted HT.

It was found that the conversion ratio of HTO from HT was clearly controlled by temperature and almost all HT was oxidized at 550 K. In addition, the gas flow rate clearly controls the conversion rate, too. The reaction rate constant above 473 K could be expressed by the following equation, $k [s^{-1}] = 5.4 \times 10^2 \exp(-0.65 \text{ eV} / k_B T)$, indicating the oxidation of tritium was governed by one elementary process.

2B04 Aggregation of actinide ion associated with electrode reduction

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(Japan Atomic Energy Agency)

Reduction of uranyl(VI) ion in weakly acidic solution and aggregation associated with reduction were investigated based on electrochemical measurements. Stripping voltammetry with a gold micro-electrode was performed after preelectrolysis. In the case of concentrated U solution of pH 3, decrease of the reduction current of U(VI) to U(V) was observed. This phenomena was explained by the deposition of reduction species of U(VI). Current peak due to oxidation of deposits was observed at +0.2 V vs SSE. These results show that deposition of reduced U on the electrode surface was very severe. In the case of pH 2, small oxidation current peak was observed at +0.2 V, but inhibition of reduction of U(VI) was not observed. On the other hand, the current peak due to deposits was appeared at -0.05 V in the case of pH 1. This fact indicates that the chemical species formed in the solution of pH 1

was different from those in lower acidity.

2B05 Systematic difference in the stabilities of complexes of hard cations with various oxygen-donor ligands

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Complexation of metal cations by ligands such as hydroxide ion, carbonate ion, carboxylate ion, and phosphate ion is one of the most important factors to control behavior of metal ions in natural environment, especially for hard ions such as actinide and lanthanide ions. Previous studies showed that these ligands were classified as intermediate hard ligands having oxygen donor, which favors to form ionic bonding. However, it was found that the reactivity of each ligand greatly depends on ionic radius and that there is a great difference of the reactivity between hydroxide ion and other ligand such as carbonate and carboxylate. However, quantitative discussion on the selectivity of metal cations is not quantitatively performed including discussion on thermodynamic parameters (ΔG , ΔH , ΔS) and quantum chemical calculation. As a result, we found that the entropic contribution to the free energy was large in the case of hydroxide complex of small radius cation. In the aqueous complexation reaction, entropy change is controlled by the number of water molecules replaced by the ligand, suggesting that hydroxide complex for large cation was not stable due to the small effect of dehydration.

2B06 Synthesis and spectroscopic properties of uranium-containing ionic liquids

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Photophysical properties such as UV-Vis-NIR spectroscopy and time-resolved laser-induced fluorescence spectroscopy of uranium(IV) and uranium(VI) complexes in ionic liquids are studied as well as synthesis and purification of uranium tetrahalides and related materials. For example, the powder of U_3O_8 was put on the glass boat under Ar atmosphere in an open glassware. To this was added CCl_4 vapor at 420 °C for two hours affording the dark green product of crude UCl_4 crystals contaminated with other uranium chlorides. Purification by sublimation was subsequently performed in a closed quartz tube at 542 °C for six hours, affording green crystals of UCl_4 . These halides were reacted with a proper amount of KSCN and 1,3-dimethylimidazolium chloride to give $[C_{2}mim]_4[U(NCS)_8]$, a target compound. There is a definitive difference in UV-Vis-NIR spectra of UX_4 dissolved in organic solvents and ionic liquids presumably due to the coordination environment around U^{4+} center. White-to-yellow luminescence of $U(NCS)_8^{4-}$ has a very short lifetime around 10 ns, which is far shorter than that of $UO_2(NCS)_5^{3-}$ species reported in our previous work with different spectral shape.

2B07 Computational approach to the selective separation of minor actinides by means of density functional theory

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(¹Grad. Sch. Sci., Hiroshima Univ., ²N-BARD, Hiroshima Univ.)

The selective separation of minor actinides (MAs) from high-level liquid waste and the disposal by the transmutation of MAs have been challenging tasks because of the difficulty of their separation. We investigated the separation behavior of MAs from lanthanides by means of

relativistic density functional calculation. For the comparison of the difference of the separation between MAs and lanthanides, we focused on the extraction behavior of MAs and lanthanides employing Cyanex-272 and -301 ligands which were phosphinic acid and dithiophosphinic acid respectively. All geometry optimization calculations were performed without any constraints for ML_3 ($M = Sm^{III}, Eu^{III}, Am^{III}$ and Cm^{III} ; $L^- = [(Me_2PO_2)_2H]^-$ and $[Me_2PS_2]^-$) complexes. All equilibrium structures at ZORA-BP86 / def2-SVP level reproduced the experimental metal-ligand lengths. Complexation energy results obtained by single-point calculations at ZORA-B2PLYP / def2-TZVP level, which gave the excellent correlation with experimental Mössbauer isomer shifts for ¹⁵¹Eu and ²³⁷Np complexes, showed the reproducibility of experimental Gibbs energy differences. This result indicated that the origin of selective separation of minor actinides is attributed to the stability of extraction complexes by the complex formation reaction.

2B08 Relations between capture cross-section measurement and prompt gamma-ray analysis

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Accurate neutron capture cross section data for minor actinides (MAs) and long-lived fission products (LLFPs) are required to estimate the production and transmutation rates for developing innovative nuclear systems. However, accurate measurements of these cross sections are very difficult due to high radioactivity of these samples. To overcome the difficulty, “Accurate Neutron-Nucleus Reaction measurement Instrument (ANNRI)” has been constructed in the Materials and Life science

experimental Facility at the Japan Proton Accelerator Research Complex. The energy-integrated neutron intensities at the flight length of 21.5 m are 1.2×10^7 n/s/cm² in the neutron energy range of 1.5-25 meV, and 1.8×10^6 n/s/cm² in 0.9-1.1 keV at a beam power of 300 kW. In the epithermal energy region, the energy-integrated neutron intensity is more than 17 times as high as the values of the other instruments. A series of neutron capture cross-section measurements with high intensity pulsed neutrons have been started.

On the other hand, not only neutron capture cross-section measurement but also prompt gamma-ray analysis with time-of-flight has been performed using the ANNRI.

In this presentation, relations between capture cross-section measurement and prompt gamma-ray analysis are presented.

2B09 Current status of development of multiple prompt gamma-ray analysis with time-of-flight method

TOH, Y.

(Japan Atomic Energy Agency)

The Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) has been designed and developed for the measurement of prompt gamma-ray analysis (PGA), nuclear cross-section data and nuclear astrophysics at BL04 of the Material and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). The germanium detector-array, which consists of two cluster-Ge detectors and eight coaxial-Ge detectors with BGO Compton-suppression shields, was installed at the flight length of 21.5m in ANNRI. The detector-array is designed to provide high gamma ray energy resolution and high detection efficiency. We have

developed a time-of-flight (TOF) prompt gamma-ray analysis (PGA) combined with gamma-ray coincidence technique. We have conducted several experiments of standard samples to assess the feasibility of the method. Neutron capture resonance peaks are clearly observed in the TOF spectra of the standard samples. This presentation will describe the detail of the experiments and the current status of the development of the method.

2B10 Elemental Analysis using Time of Flight –Multiple Prompt Gamma Ray Analysis Method on J-PARC BL04 ANNRI–

MIURA, T.¹, EBIHARA, M.², TOH Y.³, MATSUO, M.⁴, CHIBA, K.¹, KIMURA, A.³, OURA, Y.³, SHOZUGAWA, K.⁴

(¹National Metrology Institute of Japan-AIST., ²Tokyo Metropolitan Univ., ³Japan Atomic Energy Agency, ⁴Tokyo Univ.)

We are developing the new analytical method for precious metals in solid sample using the time of flight–multiple prompt gamma ray analysis method on J-PARC/MLF BL04 ANNRI (Accurate Neutron-Nucleus Measurement Instrument). The Pd-Ag alloy foil (Pd:Ag; 75:25, sample mass; 21.6 mg, thickness; 0.025 mm, 9.28 mm×9.28 mm) was selected for test sample. We presented the results and the possibility of developed analytical method.

3A01 Examination of the rapid analysis of radioactive Sr isotopes in seawater using liquid scintillation counting

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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 radio isotopes have been leaking to the underground and released into the ocean with contaminated water. Currently, for its radioactivity measurement fuming nitric acid method has been used for the analysis of ^{90}Sr in seawater. However, it is necessary to improve the method because of its time-consuming hazardous procedures. A rapid determination of radioactive Sr isotopes requires separation and purification of either of ^{90}Y and ^{90}Sr . In this study, we examined the precipitation method of ^{90}Y and the concentration method of Sr using Sr Rad-Disk. For the measurement, we examined the two techniques of the Cherenkov light measurement and the measurement using with an extraction scintillator.

3A02 Rapid determination of radiostrontium in water by solid extraction technique: part 1

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The conventional standard method used for the analysis of radioactive strontium in environmental samples is required complicated time-consuming procedure with toxic chemical. For the purpose of development of simple and quick and safety analytical methods of radioactive strontium, we used an Analig Sr-01 column (IBC Advanced Technologies, Inc.) of using a solid extraction separating agent. This method was compared with an EmporeTM Sr RadDisk (Sumitomo 3M Ltd.) of using similar agent. We applied the water samples spiked with ^{90}Sr and addition of calcium and strontium carrier. After the precipitation of calcium carbonate, the precipitate was dissolved in about

35ml solution of 2 or 3M HNO_3 . The solution was through an Analig Sr-1 column or an EmporeTM RadDisk to absorb the strontium. Activity of ^{90}Sr and ^{90}Y of the column was measured directly by well-type NaI (Horiba Ltd.). After that, Sr was eluted from the column with 0.03% EDTA solution. Eluate and disks were determined using a liquid scintillation counter LSC-6100 (Hitachi Aloka Medical Ltd.). By the quantified measurements of strontium after about three weeks at the ^{90}Sr - ^{90}Y radiochemical equilibrium, the elution rates and the absorption rate to the column or disks were estimated. The time required for chemical separation procedure by this method was 2 days although it took about 2 weeks in a conventional manner. In the range of the amount of calcium 1-5g (0.1-0.5% relative to 1L) in the sample, strontium absorption rate to the Analig Sr-1 column was 77%. This method is applicable to the screening analysis of seawater samples. The column used was confirmed to be reused. Simple determination of radioactive strontium became possible by this method.

3A03 Rapid determination of radiostrontium in water by solid extraction technique: part 2

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The standard methods used for the analysis of radiostrontium in environmental samples require complicated time-consuming procedure with toxic chemical. To explore a simple, rapid and safety radiostrontium analysis, we tried to use a column with a solid extraction separating agent (Analig Sr-01, IBC Advanced Technologies, Inc.). This method was compared with one

with a solid disk (Empore™ Sr RadDisk, Sumitomo 3M Ltd.) with similar agent. The measurement method is described here. ⁹⁰Sr was spiked in the water samples. After the precipitating, the precipitate was dissolved in about 35 ml solution of 2 or 3M HNO₃. The solution was through the column or the disk to absorb strontium. The activity of ⁹⁰Sr and ⁹⁰Y absorbed on the column was measured directly by a well-type NaI(Tl) detector (Horiba Ltd.). Then, strontium was eluted from the column with EDTA solution. The activity of the eluted solution was determined with the well-type NaI(Tl) detector and a liquid scintillation counter (LSC, LSC-6100, Hitachi Aloka Medical Ltd.). RadDisk was put in an LSC standard vial containing liquid scintillator, and was measured with the LSC. The time required for chemical separation procedure by this method was two days although it would take about two weeks by the conventional manner. Furthermore, the Analig column was confirmed to be reusable. Measurements by the liquid scintillation counting can be determined rapidly without waiting the radiochemical equilibrium. This method is applicable to the screening of seawater samples. A feasibility of simple determination method of radiostrontium was presented.

3A04 Investigation of selective extraction of cesium and strontium by solvent extraction

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(Tohoku Univ.)

As a serious result of the damaged vessel of the Fukushima Daiichi Nuclear Power Plant due to the 2011 Tohoku earthquake, the extensive leakage of the contaminated water has occurred which includes ¹³⁷Cs and ⁹⁰Sr, both are one of

the most important long-lived fission products. In order to remove these nuclides from the contaminated water, two systems had been installed including the cesium absorption filtration system and the Advanced Liquid Processing System (ALPS). However, due to the difficulties in the operation of such column systems, their operations have not been proved to be very effective for the intended decontamination.

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 function as extractants of cesium and strontium from acid solutions[1][2]. The diluents have inevitable 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. With the strong C-F bonds the diluents show excellence in the noninflammability, the low toxicity, and the chemical stability.

In this study, we investigated the use of the HFC-43-10mee and its azeotropes as diluents. We will present the results obtained by extractions of alkali metals and alkaline earth metals from aqueous solutions by dicyclohexano-18-crown-6-ether as an extractant in these diluents.

[1] R. A. Sachleben, et al., Sep. Sci. Technol., 32(1997)275.

[2] K. K. Gupta, et al., Solvent Extr. Ion Exch, 21(2003)53.

3A05 Fukushima-derived radionuclides in the atmosphere observed in Tsukuba and Iitate

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Since March 2012, the concentrations of ¹³⁷Cs and ¹³⁴Cs in aerosols were observed to be in the level of 10⁻⁵Bq m⁻³ at Tsukuba, about 170 km from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). Since March 20, 2012, we measured radiocesium in surface air at two points of Iitate (Sasu and Itamisawa), about 30km from FDNPP. The concentrations of ¹³⁷Cs and ¹³⁴Cs in aerosols were observed to be in the level of 10⁻⁴Bq m⁻³. Aerosols at Sasu showed a higher concentration of radiocesium during 31 March-8 to April 2012, which could have been caused by the re-suspension of soil particles including radiocesium for a strong wind. During March 10-21, 2013, a higher concentration of radiocesium was also observed in the Itamisawa aerosols, but in this case it was supposed to be due to the re-suspension of soil particles including radiocesium because an official decontamination activity took place at the site. The higher aerosol mass and concentration of soluble calcium ion in the aerosol supports this hypothesis. Aerosols at Sasu and Itamisawa showed a higher concentration of radiocesium during August 6-28 2013, which could have been caused by a radioactive plume of FDNPP which passed through this area from the back trajectory analysis and other information. The activity median aerodynamic diameter of radiocesium-bearing particles were larger than 1.2 μm.

3A06 Transport of radionuclides to soil at Fukushima Daiichi NPS, TEPCO
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(Japan Atomic Energy Agency)

Due to the accident of Fukushima Daiichi NPS, TEPCO, wide area was contaminated. TEPCO analyzed radioactivities of the soil within the NPS site and reported activity concentrations during 2011. Based on the data disclosed, transport behavior of the detected radionuclides from the fuel of damaged reactors to the soil was investigated. The concentration of radionuclide in the soil was normalized with the fuel composition after correcting decay and obtained relative value to that of ¹³⁷Cs as the transport ratio. The transport ratio calculated was almost constant for time of sampling and the ratios of isotopes correspond with each other. It was found that the transport ratio to the soil was in the order of followings:

I > Te > Cs > Ag~Sb > Mo~Ru > Ba > Sr > Nb
> Pu~Am~Cm

And the transport ratios for I, Sr and alpha emitters would be dependent on the location of sampling soil. Uranium was dominated by that of naturally occurred.

3A07 Properties and Distribution of Radioactive Caesium of the Surface Soil of Japanese Cedar Forest in Fukushima

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(Japan Atomic Energy Agency)

As part of "Long-Term Assessment of Transport of Radioactive Contaminant in the Environment of Fukushima" (F-TRACE project), JAEA have researched to predict the transportation of radioactive caesium from contaminated mountain forests to estuary and coast through dam, pond, river etc. based on survey data in Fukushima prefecture, and to implement technical information and remedial measures to prevent contamination of

decontaminated areas and domestic water supply. In the forest area, it is considered that radioactive caesium existed in the tree canopies move to the surface of the ground through litter fall, through fall and stem flow. In this study, we report properties and distribution of radioactive caesium of the surface soils at stem base of Japanese cedar in the Ogi district of Kawauchi-mura to know the influence of the stem flow on soil.

3A08 Fluvial discharges of particulate and dissolved radiocesium from a forest catchment

TAKEUCHI, E., ATARASHI-ANDOH, M., NISHIMURA, S., NAKANISHI, T., TSUDUKI, K., KOARASHI, J., MATSUNAGA, T.

(Japan Atomic Energy Agency)

A large amount of radiocesium emitted from the Fukushima Daiichi nuclear power plant (FDNPP) was deposited in mountainous areas. It is therefore important to determine the quantity of radiocesium transported from these mountainous areas by hydrological processes to predict the future contamination status of downstream areas. We developed an efficient system for the collection of radiocesium in both the dissolved and suspended solid (SS) forms in stream water, which comprised sequentially connected filter vessels, containing long depth-type filters (pore size: 100 and 0.5 μm), and columns filled with an absorbent to collect dissolved radiocesium. The system was applied to a small stream located about 65 km southwest from FDNPP, and the filters and columns were collected monthly. The SS samples were collected separated into four particle sizes fractions (F1: > 2000 μm , F2: 500-2000 μm , F3: 75-500 μm , F4: < 75 μm) and the radiocesium content was measured using the Ge

semiconductor detectors. The discharge ratio of particulate radiocesium increased in summer during the rainy and typhoon seasons. The dissolved radiocesium ratio reached the maximum of 46% in winter, with a decrease in the flow rate. The largest discharge of particulate radiocesium occurred in the finest size component (F4) and, as a consequence, the F4 had the highest radiocesium load.

3A09 Influence of potassium and ammonium ion in filtering the environmental radiocesium

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The potassium fertilizer is encouraged to apply for paddy fields for contamination control of radiocesium, because potassium is effective to reduce the migration of radiocesium from radiocesium-soil to crops. Prussian blue is effective to collect of cesium. The nonwoven-fabric filter containing Prussian blue (PB-filter) is able to reducing the volume by burnout. Therefore, the local government unit is considering to use PB-filter for decontaminating water which generated by decontamination of residential district. In our previous study, we have reported that NH_4^+ and K^+ ion promoted to extract radiocesium from the radiocesium-contaminated soil to water phase while these ion interfere the collection by PB-filter. In this study, to clarify the applicability to the collecting environmental contamination of radiocesium, the collecting effect of radiocesium which extracted from wood humus (*Quercus serrata*) under the conditions presence of NH_4^+ and K^+ ion by PB-filter were evaluated. Almost of radiocesium

extracted from wood was collected by twice of filtering as with Cs⁺ ion in ¹³⁷CsCl standard solution.

3A10 Elucidation of migration behavior of radioactive cesium and tritium in the tea plants

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Various radionuclides could be released by the severe accident of nuclear power plants. In particular, radiocesium (¹³⁷Cs) and tritium (³H) that have long half-lives will have a large impact on environment for a longer time. Green tea is one of special products in Shizuoka, and it is important to understand ¹³⁷Cs and ³H distribution behavior in the tea plants from the viewpoint of safety. In this study, ¹³⁷Cs and ³H distributions in the tea plants grown by ¹³⁷Cs or ³H contaminated water were evaluated by Imaging Plate and NaI (Tl) scintillation detector. ¹³⁷Cs dissolution behavior by brewing tea leaves was also estimated. ¹³⁷Cs in the tea plants grown by ¹³⁷Cs solution was localized at stems and sprouts that cell division is usually done. These results indicate that ¹³⁷Cs is chemically trapped and locally accumulated in the tea plants. In this presentation, ³H migration behavior will be also discussed.

3A11 Retention characteristics of radiocesium in lichens and their applicability as fall-out indicator

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(¹JAEA, ²TNS)

Lichens are symbiotic organisms consisted of

fungi and algae. A number of studies were carried out after the nuclear weapons tests and Chernobyl accident, and demonstrated that lichens were useful for indicator of radioactive fallout because they could take up large amount of radionuclides directly from their thallus due to lack of root system and retain them. It is essential to evaluate retention characteristics of radiocesium in lichens for the purpose of prediction of the initial concentration brought by fall-out. Especially, the retention behavior of radiocesium in lichens in Japan has not been investigated so far.

The objective of this study is to investigate the temporal change of radiocesium concentration in individual lichen. Lichen samples were parmelioid lichens grown on tree trunk of *Prunus spp.* within Fukushima prefecture. The counting rate of surface of lichen was periodically measured by means of Geiger-Mueller (GM) survey meter (ALOKA, TGS-146B) with a lead collimator to evaluate the evolution of radiocesium concentration.

The counting rate of lichens increased with the elapse of time for one year. It can be considered that lichen may take up radiocesium from stem flow including radiocesium desorbed from canopy or stem.

3A12 Distribution of radiocesium and its relationship with the other elements in tree body

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In the present work, we investigated the distribution of radiocesium and its relationship with the other elements in different parts of tree body. The primary purposes are to clarify the

uptake pathway of ^{137}Cs and to predict its behavior and accumulation within tree body. Two tree species were selected for study: Sugi (*Cryptomeria japonica*) and Koshiabura (*Eleutherococcus sciadophylloides*). Trees were collected from forests located about 30 km away from the Nuclear Power Plant. Stem disks from several vertical positions, branches and leaves were sampled. Disks were separated into bark, sapwood and heartwood. In the case of *E. sciadophylloides*, the correlation between ^{137}Cs and the other elements varied among different tree components, the correlation between ^{137}Cs and K, Rb was in the order of wood>leaves>bark, the same tendency was also observed for ^{133}Cs which had the highest correlation with ^{137}Cs compared to other elements, indicates that ^{137}Cs was well mixed with ^{133}Cs within the biological cycle in the forest ecosystems. High correlation between Mn and ^{137}Cs was only found in leaves. The chemical similarities between ^{137}Cs and K lead to the high correlation between them whereas the difference in wood and leaves may be attributable to their different physiological role.

3A13 Transfer of radiocesium in vegetables

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The accident occurred at Fukushima Daiichi Nuclear Power Plant, brought the serious radioactive material pollution to the farmland. Immediately after the accident, some vegetables exceeded the regulation value of radiocesium by direct contamination. However, quite a few vegetables grown after the accident exceeded the reregulation value of radiocesium. Absorption of radiocesium in vegetables was extremely low in

the cultivation experiments using soil contaminated by radiocesium collected in Fukushima. Absorption of radiocesium was also highly related to exchangeable potassium in soil.

Vegetables happened to be contaminated through the use of contaminated cultivation materials, such as plastic film. Application of contaminated leaf litters or hums to the soil was also influenced to the absorption of radiocesium in vegetables.

These results indicated that high radiocesium values in vegetables were mainly caused by the direct contamination due to the dissolved radiocesium from the contaminated cultivation materials.

3A14 Concentration Differences between Radiocesium and Potassium in Tissues of Herbaceous Plants

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The fate of cesium (Cs) in plant body is known as similar to that of potassium (K). However, due to their different chemical characteristics, their fates would not be completely the same, although their major chemical forms in plants are Cs^+ and K^+ . In this study, we used four herbaceous plants (giant butterbur, Japanese knotweed, tall goldenrod, and field horsetail) to see whether ^{137}Cs and ^{40}K concentrations were different among plant parts, that is, leaf blades and petioles, or leaves and stems. All samples were grown on the NIRS campus in Chiba City; after their collection, samples were separated into each parts, oven-dried and measured ^{137}Cs and ^{40}K concentrations with a Ge detecting systems (Seiko EG&G). Giant butterbur samples have been collected regularly since October 2011 (17 times in total), and every time we found that

^{137}Cs concentration in leaf blades was higher than that in petioles, while ^{40}K concentration in leaf blades were lower than that in petioles. The results indicated that distribution patterns of Cs and K were different in different plant parts. Similar tendency was also found in leaves and stems of other plant samples.

3A15 Correction of Sum coincidence effect using total efficiency

- Sum Coincidence effect correction for Ag110m -

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(Kanazawa Univ., LLRL)

The correction of sum coincidence effect would require for nuclide with cascade gamma-ray emission, such as Cs134, Al26, Ag110m, Na22 and Co60. The correction factors of these nuclides have not been estimated from peak to total ratio, but from total efficiency as a function of gamma-ray energy. The total efficiency curve as a function of energy leads to correction of sum coincidence effect of all of nuclides including those with extremely complicate decay scheme, such as Ag110m. Correction of sum coincidence effect by total efficiency is practical.

3A16 Development of the modified sum-peak method

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The sum-peak method is one of the absolute radioactivity measurement methods, which is applicable to radionuclides emitting more than one photon in coincidence. The method requires

the knowledge of the total count rate as well as the peak count rates and the sum peak count rate. However when a sample contains other radionuclides, it is fraught with difficulty to evaluate the total count rate of the corresponding radionuclide. To solve the problem, we modified the formula of the method by removing the total count rate and tried to evaluate the activity. The modified sum-peak method was theoretically investigated and experimentally verified with two HP-Ge detectors using ^{60}Co , ^{22}Na , and was successfully confirmed. Although the accuracy is limited, it was proved that the modified sum-peak method was effective to measure samples containing multiple radionuclides and was quite simple and practical.

3A17 Application of the sum-peak method to ^{134}Cs activity measurement

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^{134}Cs emits more than ten gamma-rays per a single decay, so the decay scheme is quite complex. Therefore, there are few reports about the activity determination of ^{134}Cs by the sum-peak method. In this study, the conventional sum-peak method and the modified sum-peak method developed by us are applied to determination of ^{134}Cs activity. It was successfully proved that both the methods were applicable to determination of the activity. Especially, the modified sum-peak method was useful to determine the activity of the sample containing other radionuclides; such as ^{137}Cs . Furthermore, the modified sum-peak method potentially has an ability to determine the

activity of a volume sample with nonuniform distribution of activity. Consequently, it was proved that the modified sum-peak methods were quite effective to measure samples extracted from Fukushima.

3B01 Validation of matrix correction models in quantitative EPMA for accurate Chemical U-Th-total Pb Isochron Method (CHIME) dating

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The CHIME dating is a chemical dating method using U-Th-Pb series. This method provides micro-volume subgrain dating using electron probe micro analysis. Accuracy of the CHIME dating depends of the quality of chosen matrix model in addition to that of X-ray measurement and standard materials because an age is obtained from a set of chemical compositions of U, Th and Pb. The surface-center Gaussian model by Armstrong[1] with the atomic number correction by Love et al.[2] gives inaccurate chemical compositions of U, Th and Pb, while the double parabola model by Pouchou & Pichoir[3] gives accurate chemical compositions. It is result of (1) mathematical limitation in the surface-center Gaussian model and (2) inaccuracy of electron backscattering correction by Love et al.[2]

[1] Armstrong, J.T. (1991) In: Electron Probe Quantitation, Heinrich, K.F.J. & Newbury, D.E. (eds), Plenum Press, New York, 261 – 315.

[2] Love, G., Cox, M.G. & Scott, V.D. (1978), J. Phys. D, 11, 7 – 21.

[3] Pouchou, J.L. & Pichoir, F. (1991) In: Electron Probe Quantitation, Heinrich, K.F.J. & Newbury, D.E. (eds), Plenum Press, New York, 31 – 75.

3B02 The indirect radiocarbon dating using microscope observation and bibliographical consideration of kohitsugire

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Ganjin brought Japan many Buddhist sutra, Buddha statues, medicine and spice. Especially an important thing is the Shibunritsu sutra of 60 volumes which described commandments and organizational operation of temple. At present, the Shibunritsu sutra of 16 volumes that are said for Ganjin to have brought are stored in Shosoin. Radiocarbon dating is destructive analysis. It, therefore, cannot be applied to Sutra stored in Shosoin directly. Kohitsugire is a fragment of ancient calligraphy. They were cut from an ancient book or roll of sutras. We obtained a kohitsugire considered to have been cut from Shibunritsu stored in Shosoin. Microscope observation and bibliographical consideration clarified that the kohitsugire is rightly fragment of the Shibunritsu stored in Shosoin. Radiocarbon dating of the kohitsugire indicated that it was written before the Ganjin visit to Japan. This result shows that possibility that the kohitsugire and Shibunritsu of Shosoin were brought about by is very high. This study using microscope observation, bibliographical consideration and radiocarbon dating indicated that indirectly radiocarbon dating can be applied for samples to which destructive analysis could not be originally applied.

3B03 Radiocarbon dating of carbonized material attached to pottery: implications of chemical component of inner and outer

surfaces on potsherds

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The difference of radiocarbon ages are examined for the charred materials between inner and outer surfaces on pottery from a wetland archaeological site, and discussed in light of chemical component on the carbonized materials. The samples consist of eight pieces of potsherds, which are excavated from Irienaiko archeological site, around the lake Biwa, Shiga prefecture, Japan and are found to be originated from the same pottery of the “Kitashirakawa-kasou IIc type” in the Early Jomon era. The AMS radiocarbon dating for these potsherds indicates that the inner surface (charred food residues) of the potsherds (5052 ± 12 BP; N = 5) are about 91 ± 25 ¹⁴C year older than that of outer (soots) ones (4961 ± 22 BP; N = 7; except for SGMB-4232b). There is no marine reservoir effect of the samples from Irienaiko site, because the $\delta^{13}\text{C}$ values of inner charred matters after AAA treatment are less than -26‰ notation, which means typical of terrestrial origin. Therefore, the difference of ages is likely to be caused by freshwater reservoir effect, the ages of outer surface (soots) were corresponded to the age of the site.

The chemical composition of outer surface sample SGMB-4232b, which was from the outer surface, was similar to that of the other outer surface samples, but the radiocarbon age of this sample was similar to those of the inner surface samples after AAA treatment. Before AAA treatment, the chemical composition of

SGMB-4232b was similar to that of the inner ones. There was a possibility that some of “food residues” were attached to the outer surface on pottery and burned out. The origin of “boiling over” in outer surface on pottery could be possibly explained by chemical compositions of SGMB-4232b.

In this paper, we also discuss the correlation of the radiocarbon ages found in inner and outer surfaces on potsherds, with the change of chemical compositions (C/N ratios) before and after AAA treatment and the difference of lipid composition in inner and outer surfaces on potshers, which may provide a scientifically evidence to explain “boiling over” in outer surface on pottery.

3B04 Determination of V in geological reference samples by INAA and ICP-AES

TAKAHASHI, H., SHIRAI, N., EBIHARA, M. (Tokyo Metropolitan University)

Vanadium is widely distributed in the Earth’s crust and mantle. It is geochemically classified into lithophile elements. The determination of V for cosmochemical and geological rock samples is important in unraveling early history of our solar system and evolution of planetary bodies respectively. Several analytical methods (ICP-AES, ICP-MS, INAA, AAS, and XRF) have been developed for the determination of V at low concentration level. Among them, ICP-AES is the most frequently used nowadays. In ICP-AES, several wavelengths were used in previous studies, but a suitable wavelength for the determination of V has not been examined. In new study, we valued three wavelengths (292.402nm, 310.230nm and 311.071nm) by analyzing geological reference samples and comparing ICP-AES data with INAA data. It was observed that 292.402nm and 311.071nm

have spectral interference from Ti and that interference at 311.071nm is 10 times higher than that at 292.402nm. Argon gives spectral interferences for 310.230nm, increasing the background higher than those for other wavelengths. Considering detection limits and spectral interferences, we concluded that V could be accurately determined by using the 292.402nm emission line in ICP-AES.

3B05 Multielemental analysis of NIST and IAEA reference materials (NIST-1646a, NIST-1400, IAEA-450 and IAEA-395) by INAA, ICP-AES and ICP-MS

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(¹Tokyo Metropolitan Univ., ²Philippine Nuclear Research Inst., ³Nuclear Research Inst.)

Certified reference materials (CRMs) are greatly important in chemical analysis. They can be used as reference standard or quality control samples. A survey of literatures reveals that the number of published values for NIST-1400 (Bone Ash), supplied from the Institute of Science and Technology (NIST), and two of IAEA-450 (Algae) and IAEA-395 (Urban Dust) supplied from International Atomic Energy Agency (IAEA), is very limited. In this study, the multi-element abundances in these three CRMs were determined. A combination of three standardized analytical methods involving instrumental neutron activation analysis (INAA), inductively coupled plasma atomic emission spectroscopy (ICP-AES), and the inductively coupled plasma mass spectrometry (ICP-MS) has been applied to obtain a wide range of elemental abundances in the mentioned samples. Our analytical data was evaluated by analyzing NIST-1646a Estuarine Sediment. Our data for NIST-1400, IAEA-450 and IAEA-395 are in

good agreement with certified and reference values. Although absolute REE abundances for NIST-1400 and IAEA-450 are different, CI-normalized REE abundances patterns for these three reference materials are similar to each other and continental crustal materials. Through this research, a new data set of twenty seven, 28 and 39 elements for NIST-1400, IAEA-450 and IAEA-395, respectively, have been proposed.

3B06 Neutron activation analysis of household aluminum foils – Comparison between comparative method and k_0 -IAEA method –

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(Tokyo Metropolitan Univ.)

Instrumental neutron activation analysis (INAA) using the k_0 method can determine all elements that emit γ rays as it does not require a reference standard sample. However, some parameters, such as a neutron characteristics and an efficiency of a detector, should be determined prior to an elemental determination. In this study, household Al foils were analyzed by k_0 -INAA to evaluate impurity concentrations and their determination values were compared with those by a comparative method. Household Al foils are often used in the cooking, and recently cheap imported foils are marketed. Thus impurities in Al foils are interesting. To evaluate the determination values by the k_0 -INAA method, some reference materials were also analyzed. Elemental compositions obtained by k_0 -INAA method were fairly consistent within 10% with those of literature values for most of elements. For Al foil samples, V and Mn were determined from irradiation for ten seconds. Those determination values by k_0 -INAA were systematically about 10% smaller than those by

a comparative method. The concentrations of V and Mn in Al foils varied from 25 to 300 ppm and from 14 to 220 ppm, respectively. It was found that the amount of impurities vary greatly according to the manufactures.

3B07 INAA of Chinese medicine and herbs

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At the APSORC13 in 2013, INAA results of Chinese medicine and herbs were reported. The Niu Huang Jiedu Pian and the Liushen Wan, which were purchased with a doctor prescription in China, added to As and Hg with extremely high concentration. Five kinds of the same named Chinese medicine were gotten by a personal import with the Internet and analyzed by neutron activation and X-ray Diffraction (XRD) method. It was clear that both of the As and Hg were included in these samples by INAA and their chemical forms were identified as sulfide by the XRD analysis. It is considered that these minerals of realgar and cinnabar were added to some of these medicines named Niu Huang Jiedu Pian and Liushen Wan. Also it is considered that these sulfides work to give a shock to the body without absorption of them. Other Chinese medicine and herbs were analyzed and many kinds of elements were detected in them, which were main elements of the crust.

3B08 Factor Affecting Activity Distribution of Radioactive Aerosols Formed in Accelerator Target Room

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In accelerator facilities, particle size

distributions of radioactive aerosols are an important factor to estimate occupational internal dose. In this study, the number size distributions of aerosols formed in the accelerator target room of KUR LINAC were measured by a scanning mobility particle sizer (SMPS). Formed aerosols were also collected by a low-pressure cascade impactor and activity of aerosols collected on collection plates was measured by a GM counter. In the target room, a Ta target was irradiated with 30 MeV electron beam and currents of 20-100 μ A for about two hours. The number of aerosols increased with beam current. However, the count median diameters of the number size distribution was approximately constant without depending on beam currents. Under the irradiation condition with a current of 80 μ A, the median diameter of activity size distribution of aerosols collected by the impactor was about 1.3 times as large as that estimated from the number size distribution measured by the SMPS. This would be because that small particles or gases attached to collection plates of the impactor.

3B09 Environmental factors on ⁹⁰Sr concentration in shallow well water

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In the course of environmental radiation monitoring around RRP and related facilities, the concentration of ⁹⁰Sr in water of one well gradually increased from 3mBq/L in 2003 and reached 28mBq/L in 2011. Considering the specific activity of ⁹⁰Sr and operation history of RRP, this phenomenon was attributed to fallout from past nuclear tests. For further understanding, we studied from various aspects.

Results of the study were summarized as following. 1) Specific activity of ^{90}Sr stayed constant none the less of increase of concentration of ^{90}Sr . 2) Concentration of ^{90}Sr correlated with electric conductivity (EC). 3) EC value rapidly changed in short term. 4) Concentrations of sodium and chloride showed better correlation with EC. 5) Small surface stream near the well connected with small sand basin, whose water level was influenced by snow dumping.

Thus, following mechanisms were suggested. 1) Concentration of ^{90}Sr in the well water changed in accordance with non-radioactive substances. 2) The well water quality was a mixture of some components. 3) Land development and snow dumping might be reasons for gradual change of well water quality. We concluded that the quality of the shallow well water was easily affected by local condition and human activities.

3B10 Application of ICP-MS to determination of ^{129}I in field soil around Tokai Reprocessing Plant

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(Japan Atomic Energy Agency)

Iodine-129 (^{129}I) has been discharged into the environment from the nuclear weapons tests, nuclear power plant accidents, and nuclear fuel reprocessing plants. Therefore, the distribution of ^{129}I in field soil around Tokai Reprocessing Plant has been studied since 1982 by Japan Atomic Energy Agency. Neutron activation analysis and accelerator mass spectrometry (AMS) have been used for the determination of ^{129}I . However, both methods depend on the operation schedule of the facility. In this study, we examined the possibility to determine ^{129}I in

field soil samples collected around Tokai Reprocessing Plant by triple quadrupole inductively coupled plasma mass spectrometry (ICP-QQQ). At first, we found the best measurement condition of ICP-QQQ for reducing the background intensity of m/z 129 by ^{129}Xe included in plasma gas and polyatomic ions such as $^{127}\text{IH}_2^+$. Iodine-129 concentrations in the field soils were then measured by the optimized ICP-QQQ protocol after isolating iodine by combustion-solvent extraction method. The results of ^{129}I concentrations agreed with the values obtained by AMS. This suggested the applicability of ICP-QQQ to determination of ^{129}I in field soil around Tokai Reprocessing Plant.

3B11 Sequential separation of ultra-trace multi-elements including Pu

MIYAMOTO, Y., YASUDA, K., MAGARA, M. (Research Group for Analytical Chemistry, JAEA)

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, and the lanthanides using a single anion-exchange column and mixed media consisting of hydrochloric acid, nitric acid, acetic acid, and hydrofluoric acid in the previous work. The chemical composition of eluents for separation of Pu and Th was optimized to accomplish the sequential separation of multi-elements including Pu. The mixture of 11 pg of Pu and 200 pg of Th was completely separated, and >95% of these

elements were recovered by a single anion exchange column with the mixed-media of 70 vol.% acetic acid, 2 M HCl, and 0.1 M HF. By using this mixed-media for separating Th and Pu, the sequential separation of ultra-trace multi-elements including Pu was accomplished.

This work was supported by financial aids from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan (Scientific Research (C): Grand No. 25340078).

3B12 Alpha-ray measurement of dissolved plutonium particle on the TIMS filament

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(Japan Atomic Energy Agency)

In order to detect of undeclared activities in nuclear facilities, we develop to technique of safeguards environmental sample analysis in JAEA. As part of this effort, isotope ratio analysis of individual MOX particle was performed using by TIMS without chemical treatment. It is possible to estimate an age determination of plutonium purification by a combination of the plutonium isotope ratios and $(\text{Pu-238}+\text{Am-241})/(\text{Pu-239}+\text{Pu-240})$ alpha activity ratios were obtained by the alpha spectrometry. Since the plutonium particle size is more than 1 μm due to the influence of self-absorption, exactly measurement of the activity ratios has been difficult. Therefore, we were examining how to acid dissolution in the plutonium particles on the TIMS filament before alpha spectrometry.

3B13 EEC radon in underground research facilities

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(JAEA)

In the Mizunami Underground Research Laboratory of Japan Atomic Energy Agency, horizontal tunnels and vertical shafts are excavated, and many kinds of observation of underground environment are performed as geoscientific research related to the geological disposal technology of high-level radioactive waste. Since the geological feature of this area mainly consists of granite, measurements of the radon equilibrium equivalent concentration (EECRn) in the underground research facilities and dose evaluation to workers were performed. EECRn was measured intermittently using an instrument WLM-Plus (Tracerlab Co., Ltd.) in 2012 and 2013. The measurements were performed at two locations, where relatively high EECRn values were observed before, at a depth of 300m below the surface. Trends to increase EECRn during summer as a seasonal variation were observed, because the natural ventilation of underground facilities was suppressed by high air temperature outside. The maximum value of 3,300 Bq/m was measured in July 2013. During the workers entrance time zone, 10:00 to 15:00 when ventilation blowers were operated, the maximum EECRn of 1,880 Bq/m was observed in July 2013 and the monthly maximum average value was 765 Bq/m in August 2013. Using the monthly average EECRn, the annual effective dose to workers was estimated to be about 0.35 mSv on the assumption of 10 hours working time at the area per each month.

3B14 North-south transection of Be-10 concentration in seawater in the Indian Ocean

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Cosmogenic nuclide Be-10 (half-life 1.36 Ma) is produced by nuclear interaction between secondary cosmic ray and atmospheric nitrogen and oxygen. Because of Be-10 widely spread in the ocean and the residence time is shorter than ocean-mixing time, the Be-10 concentrations in seawater could be a useful tracer for the ocean circulation. It is necessary accumulation of oceanic Be-10 to assess Be-10 as a tracer for water mass. In this presentation, the north-south transection of Be-10 concentration shows in Indian Ocean during R/V Hakuho-maru in the KH09-5 cruise, and discuss a migration of Be-10.

The concentration of Be-10 is high in 20-40°S and low in the south area of 40°S, which is the Antarctic Ocean, and between 20°S and 20°N in surface. According to hydrological data, Be-10 concentration in Antarctic intermediate water (AAIW) is relatively low concentration. The AAIW come from the low concentration surface water of the Antarctic Ocean. In deep-sea, the water mass, which named Common deep water, has high concentration. The Common deep water is originated from Atlantic deep sea, but the concentration of Be-10 in the Atlantic Ocean is lower than Common deep water. It is consider that the Be-10 in the surface Antarctic Ocean is transported down to the Antarctic deep sea by biogenic particles in Antarctic Ocean.

3B15 Application of pulsed optically stimulated luminescence using natural mineral to retrospective dosimetry

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(¹Japan Atomic Energy Agency)

Optically stimulated luminescence (OSL)

from natural quartz and feldspar has been used in retrospective dosimetry, dating of sediment, etc. In the application target minerals must be extracted with chemical separation which requires cumbersome work. Nowadays, pulsed optically stimulated luminescence (P-OSL) was developed to separate luminescence signal from between quartz and feldspar as instrumental separation. The purpose of this study is to try apply the P-OSL to Japanese surface soil as retrospective dosimetry with easy physical separation.

In this study, a P-OSL instrument was developed by the author. Performance of the instrument was tested using natural quartz extracted from Japanese surface soil and feldspar in mineral specimens. By using a mixture of the quartz and feldspar, the P-OSL protocol was improved to determine the irradiation dose. Following irradiation using X-rays, dose recovery test was conducted and the irradiated dose was successfully reconstructed by the P-OSL protocol. Moreover, as-received surface soils were irradiated with Co-60 source of the Research Reactor Institute, Kyoto University and used in the dose recovery test. The test was also successful in reconstructing the irradiated dose. From these results, it was concluded that the P-OSL dosimetry is suitable for accident evaluation although further research is required to improve the protocol using various kinds of Japanese surface soils.

3B16 Study on tardigrades with radiation resistance

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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. Many tardigrades live in terrestrial mosses and lichens. The bodies of tardigrades are short, plump, and ventrally there are four pairs of stubby legs and each leg have four to eight claws.

Tardigrades exhibit remarkable ability to withstand for extreme environment. When the mosses become desiccated the tardigrades contracted and shriveled, and pass into an anabiotic state. An animal in such a condition is called a tun. When water is again present, the animal swells and becomes active within a few minutes. Tardigrades recover after immersion from tun.

We investigated the effects of environmental stress on tardigrades (*Milnesium tardigradum*), such as examined the effects of high or low temperature, and gamma-rays. Especially we examined the effects of gamma-ray irradiation equipment in Radioscience Research Laboratory, Faculty of Science, Shizuoka University.

We found that tardigrades withstand high or low temperature, and 4000 Gy gamma-rays. This result suggests that tardigrades have a great ability to withstand radiation than other organism.

P01 Study on solvent extraction behavior of zirconium and hafnium as homologs of rutherfordium (element 104) with chelate extractant

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Chemical studies on rutherfordium (Rf) have been carried out mainly with inorganic ligands

such as halide ions. However, little has been reported on chemical experiments of Rf with organic ligands. In this study, we carried out the solvent extraction of zirconium (Zr) and hafnium (Hf) as lighter homologs of Rf using 2-thenoyltrifluoroacetone (TTA) as a chelate extractant toward the Rf experiment. In this experiment, radiotracers ⁸⁸Zr and ¹⁷⁵Hf were used. These radiotracers were produced in the ⁸⁹Y (p, 2n) and ¹⁷⁵Lu (p, n) reaction, respectively, using the AVF cyclotron at Tohoku University. Aqueous solution of 1 M HNO₃ containing the radiotracers was mixed with an equal volume of TTA in toluene solution, and the mixture was then shaken for 5 hours. The radioactivity of each phase was measured by a Ge detector. The results showed that the distribution ratio (*D*) of Zr is approximately 10 times greater than that of Hf. The log *D* vs. log [TTA] plot indicated that the extracted TTA complexes of Zr and Hf presumably contained OH⁻ or NO₃⁻ ion as ligands in this experimental condition. In the symposium, we will also report on the results of solvent extraction from perchloric acid solution.

P02 Solid-liquid extraction of Zr and Hf using automated solid-liquid extraction apparatus for element 104, Rf, experiment

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Solid-liquid extraction of Zr and Hf, homologues of Rf (*Z* = 104), by Aliquat 336 resin from hydrochloric acid (HCl) were

performed using automated batch-type solid-liquid extraction apparatus for repetitive extraction experiment on Rf. The Zr and Hf species adsorbed on the resin in 10 M HCl were stripped by washing the resin several times with ~0.25 mL of 4 M HCl. The back extraction yields of Zr were higher than those of Hf. The back extraction yields of Zr and Hf decreased with an increase of the Aliquat 336 concentration. Under these conditions, constant distribution coefficients (K_d) were obtained in repetitive ~100 times extraction experiments of Zr and Hf without renewing the resin. Using this apparatus, we performed solid-liquid extraction employing $^{89\text{m,g}}\text{Zr}$ and $^{169,173}\text{Hf}$ produced by nuclear reactions and transported continuously from the nuclear reaction chamber by the He/KCl gas-jet system at RCNP and RIKEN. The dependences of the K_d values of Zr and Hf on the shaking time were obtained in the online experiment in 9 M HCl. The K_d values in online experiment were in agreement with those in the offline experiment.

P03 Measurements of cross sections for production of light nuclides by 120 GeV and 400 MeV proton bombardment of Y

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By accelerator mass spectrometry (AMS), we have measured the ^{10}Be and ^{26}Al production cross sections from Y, Tb produced by 120 GeV and 400 MeV protons. The proton irradiation at 120 GeV and 400 MeV were performed at Fermi National Accelerator Laboratory (FNAL) and the Research Center for Nuclear Physics (RCNP), Osaka University, respectively. The AMS measurements were performed at MALT (University of Tokyo). We will present the correlation between the cross section data of ^{10}Be and ^{26}Al and the target mass number and discuss the target mass dependence in those cross sections.

P04 Isothermal chromatographic behavior of BiCl₃

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In the study of super heavy elements, a technique of an isothermal chromatography has been often used to determine adsorption enthalpies of volatile compounds. The results of such experiments have been analyzed mostly using Monte Carlo method. In this simulation, experimental parameters such as a column length and radius are necessary but they have more or less uncertainty. Therefore it is necessary to ascertain an accuracy of adsorption enthalpy for a typical material. In this study, we consider whether volatile BiCl₃ can be used as a typical material, because we can easily handle ^{212}Bi as a tracer from the naturally occurring radioactive series. First of all, we have

investigated isothermal chromatographic behavior of BiCl_3 in macro-scale. BiCl_3 is deliquescent and changes to nonvolatile BiOCl when rapidly heated. So, it needs to dehydrate BiCl_3 by gradual heating at 100°C while flushing He gas. In this experiment, BiCl_3 was heated at 100°C for 1 hour with He flow (0.8 L/min) at atmospheric inner pressure. Then, the temperature was raised at 500°C and volatile compound was transported to a collection position. The amount of collected Bi was determined with spectrophotometry using thiourea. The result was that the quantity of collected BiCl_3 reached maximum at between 1 and 3 min. It was found that BiCl_3 started to evaporate from $\sim 250^\circ\text{C}$ immediately.

P05 Summary on the 113th element search using GARIS

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During ten years from 2003 to 2012, we have studied on a production and decay properties of a new superheavy nuclide $^{278}113$ produced by $^{209}\text{Bi}(^{70}\text{Zn},n)^{278}113$ using a gas-filled recoil separator GARIS. Totally 3 decay chains due to the $^{278}113$ were observed during net irradiation time of 576 days. The 1st and 2nd decay chains consist of four alpha decays from $^{278}113$ to ^{266}Bh , and then ended by spontaneous fission of ^{262}Db . The 3rd chain was consist of 6 alpha decays and connected to ^{254}Md . □ Observed decay properties from 3 decay chains were consistent each other. In 2009, we also tried to search for the ^{266}Bh produced by $^{248}\text{Cm}(^{23}\text{Na},5n)^{266}\text{Bh}$ to establish the connection to known nuclides and the cross-bombardment. As a result, totally 14 events were assigned to decay chains from

^{266}Bh . □ The identification was based on a genetic link to the known daughter nucleus ^{262}Db by α -decay. Thus, the decay chains from ^{266}Bh were well established. Therefore, we concluded the 3 decay chains from $^{278}113$ are clearly connected to the anchor nuclides and the productions of $^{278}113$ are clearly confirmed.

P06 Excitation functions for the $^{\text{nat}}\text{Hf}(\alpha,x)$ reactions

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We previously investigated the production of a long-lived Ta isotope, ^{179}Ta ($T_{1/2} = 1.82$ y), in the $^{\text{nat}}\text{Hf}(p,x)$ and $^{\text{nat}}\text{Hf}(d,x)$ reactions for the basic chemical studies of 105th element, Db. However, ^{179}Ta emits no γ rays in its EC decay and therefore it is assayed by measuring the characteristic X rays of Hf. The isotope ^{182}Ta ($T_{1/2} = 114.74$ d) is a more useful radiotracer because it emits characteristic γ rays. In this work, we investigated the production of ^{182}Ta by the α -induced reaction on $^{\text{nat}}\text{Hf}$.

A stack of the thin foils of $^{\text{nat}}\text{Hf}$ and $^{\text{nat}}\text{Cu}$ was irradiated by the 50-MeV α beam for 31 min. The average beam current of $0.23 \mu\text{A}$ was determined by the $^{\text{nat}}\text{Cu}(\alpha,x)^{67}\text{Ga}$ monitor reaction. After the irradiation and proper cooling duration, γ rays of each foil were measured by a Ge detector.

In this work, the cross sections of the $^{\text{nat}}\text{Hf}(\alpha,x)^{176,177,178}\text{W}$, $^{175,176,177,178,182,183}\text{Ta}$, $^{175,179\text{m}2}\text{Hf}$ reactions were measured. The measured excitation function for the $^{\text{nat}}\text{Hf}(\alpha,x)^{182}\text{Ta}$ reaction exhibits the maximum cross section of 8.3 ± 0.3 mb at 41.5 ± 1.2 MeV. Based on the measured cross sections, the

thick-target yield of ^{182}Ta was deduced to be 5.2 kBq/ μAh .

P07 Measurement of production cross sections of Tc isotopes in the $^{nat}\text{Mo}(d,x)$ reactions

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We plan to conduct model experiments for chemical studies of element 107, bohrium using its homologues, Tc and Re radioisotopes. In this work, production cross sections of deuteron-induced nuclear reactions on natural molybdenum have been measured up to 24 MeV for the quantitative production of the Tc isotopes. The target stack of Mo/Ti/Ta foils was irradiated for 1 h with the 24-MeV deuteron beam supplied from the RIKEN AVF cyclotron. After the irradiation, each foil was subjected to γ -ray spectrometry. The beam current of 0.17 μA was determined by the monitor reaction $^{nat}\text{Ti}(d,x)^{48}\text{V}$. The excitation functions were measured for the $^{nat}\text{Mo}(d,x)^{93\text{m},93\text{g},94\text{m},94\text{g},95\text{m},95\text{g},96\text{m},96\text{g},97\text{m},99\text{m},101}\text{Tc}$, $^{93\text{m},99,101}\text{Mo}$, $^{90\text{g},92\text{m},95\text{m},95\text{g},96\text{g}}\text{Nb}$, $^{89\text{g}}\text{Zr}$ reactions. They were discussed by referring to previously reported data and to the theoretical model code TALYS. Most of our results are in good agreement with the previously reported data, though there is little agreement between the experimental values and the theoretical ones. Thick-target yields for the investigated isotopes were deduced from the measured production cross sections.

P08 Isothermal gas chromatographic behavior of Zr and Hf chlorides at off-line experiment

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To clarify chemical property of element 104, Rf, gas chromatographic behaviors of chlorides of Rf and its homologs, Zr and Hf, have been studied in a single-atom scale. The reported sequence of volatility was $\text{Zr} \gtrsim \text{Rf} > \text{Hf}$. But this volatile relation between Zr and Hf chlorides differs from sequence of sublimation expected from their vapor pressure curves in the macro-scale. Appropriate explanation about this difference has not been given until now. In this study, we investigated gas chromatographic behaviors of Zr and Hf chlorides at off-line to clarify this contradiction. In macro-scale, we obtained isothermal chromatographic data of Zr and Hf chlorides. With analyzing these retention curves, it was found that HfCl_4 is more volatile than ZrCl_4 , which agrees with the volatility deduced from the vapor pressure. To investigate whether this chromatographic behavior follows the model of mobile adsorption, we are going to perform the experiment of the isothermal gas chromatography experiment using radioactive tracer. In the symposium, the experimental conditions and the results will be presented in more detail.

P09 Solvent extraction behavior of Mo and W from oxalic acid solution by Aliquat 336

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In this work, ion-pair extraction behavior of molybdenum (Mo) and tungsten (W) as homologs of seaborgium (Sg) has been investigated with Aliquat 336. Radiotracers ^{93m}Mo , ^{176}W and ^{181}W were dissolved in 10^{-2} M $\text{H}_2\text{C}_2\text{O}_4$ (oxalic acid)/0.1 M HCl/0.9 M LiCl solution, and the solution was shaken with Aliquat 336/toluene solution. After separation of the aqueous and organic phases, distribution ratios (D) of Mo and W were calculated from the radioactivity in each phase measured by γ -ray spectrometry with a Ge detector. In the experiment of extraction kinetics, D values of Mo and W were constant in shaking time longer than 30 s. This result shows that the extraction kinetics of Mo and W with Aliquat 336 from oxalic acid solution is fast. Moreover, the D values of Mo and W as a function of concentration of Aliquat 336 were investigated. Slopes in the D value vs. [Aliquat 336] plot on a logarithmic scale were evaluated to be 2.1 for Mo and 1.3 for W. This indicates that the net charge of the extracted anionic oxalate complex of Mo is -2, while that of the extracted W complex is -1, suggesting that chemical species of W is different from that of Mo.

P10 Development of a collection apparatus for recoil products toward the study of the deexcitation process of U-235m

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^{235m}U has very low excitation energy (76.8 eV) and decays predominantly by the internal conversion process. The deexcitation of ^{235m}U is caused by the interaction between the nucleus and outer-shell electrons, and thus the variation of the decay constant 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 ^{235m}U with various chemical forms. In this work, we developed an apparatus for collecting ^{235m}U recoiling out of ^{239}Pu . We evaluated the performance of the apparatus by using ^{224}Ra recoiling out of ^{228}Th . The collection yields of ^{224}Ra were determined in various applied voltages, air pressures, and ^{228}Th source shapes. Based on these results, we determined suitable experimental conditions for the collection of ^{235m}U from ^{239}Pu . In addition, the detection apparatus for the low-energy internal-conversion electrons are under development.

P11 R&D of focal plane detector for superheavy element study

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Superheavy (SHE) element device development team in RIKEN has developed a new focal plane detector to study on production and decay properties of SHE nuclide using a gas-filled recoil separator GARIS and GARIS-II. Conventional identification method of SHE nuclide was based on the genetic relation between mother and daughter, was obtained from measuring the α -decay and spontaneous fission (SF) by Si detector box. So, detailed decay scheme including γ -decay could not be obtained. In 2013, we newly installed a Si-Ge detector array at the focal plane of GARIS for studying on production and decay properties of reaction products by $^{248}\text{Cm}+^{48}\text{Ca}$. The detector array is also useful as a probe for detection of

prompt γ -ray coincided with SF. Before the experiment on the $^{248}\text{Cm}+^{48}\text{Ca}$, we calibrated the Si-Ge detector array using nuclear reactions of $^{207}\text{Pb}(^{48}\text{Ca},2n)^{253}\text{No}$, $^{208}\text{Pb}(^{48}\text{Ca},xn)^{256-x}\text{No}$ [$x=1,2,3$], and $^{206}\text{Pb}(^{48}\text{Ca},2n)^{252}\text{No}$. In this report, the results on the first α - $\gamma(X)$ and SF- γ spectroscopic study at the focal plane of GARIS will be given in detail.

P12 R&D of target for superheavy element production and its monitoring system

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In order to produce a superheavy element (SHE), it is necessary a target system to stand against high intense heavy-ion irradiation. Conventionally, a thermal damage such as a melting of target material is avoided using a gas-cooled rotating target system for the heaviest element production. There are thermal conduction, convection, and radiation as a cooling process of target. It is important in target development to know the cooling mechanism and the time scale for cooling. So, we measured the target's temperature increased by heavy-ion irradiation using a radiation thermometer without contact. In this report, we will discuss about the target's cooling mechanism. Additionally, we will also introduce an on-line target monitoring system.

P13 Adsorption behavior of Zr and Hf by TTA-reversed-phase chromatography for aqueous chemistry of a super-heavy element Rf

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Studies on the fluoride complex of Rutherfordium (Rf), a super-heavy element, had been performed by a cation-exchange chromatography method in HF/HNO₃ solution. Speciation of the Rf complex is, however, still open question. We, therefore, aim to clarify cationic fluoride species of Rf in aqueous solution and to determine its formation constant. At present, we focus on the extraction with an acidic chelate extractant, TTA, sensitive to valence of the metal complex. In the present study, on-line TTA-reversed-phase chromatography and batch extraction experiments were performed using carrier-free Zr and Hf radiotracers in HF/HNO₃ solution to examine the applicability of the TTA-reversed-phase chromatography to Rf. Distribution coefficient (K_d) values of Zr and Hf in both the experiments were determined and were compared in a wide range of the fluoride ion concentration. It is found that the fluoride complexes of Zr and Hf are increasingly formed with an increase of fluoride ion concentration. The K_d values obtained in the chromatography experiment agreed well with those by the batch method at higher concentrations of fluoride ion. Besides, clear difference of K_d values between Zr and Hf was observed in the concentration range of $> 2 \times 10^{-5}$ M. These indicate that the present conditions are promising for our future Rf experiment with TTA-reversed-phase chromatography.

P14 Effect of excited states in atom and ion contributing to the surface-ionization efficiency

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To determine the first ionization potential of the heaviest actinide Lr, we have newly developed the surface ionization method which can be applied to the elements with short half-lives and very low production yields. To test the applicability of this method, we have investigated a relationship between ionization potentials and surface ionization efficiencies of various short-lived lanthanide elements. We have found that the effect of excited states in atoms and ions largely contributes to the measured ionization efficiencies of lanthanide elements.

P15 Development of an online perturbed angular correlation measurement system for the application of the short-lived $^{19}\text{F}(\leftarrow^{19}\text{O})$ probe

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It is important to obtain information on local structures surrounding fluorine (F) atoms in order to study physical properties of matter containing F atoms. For that purpose, we have adopted the perturbed angular correlation (PAC) technique, and are trying to measure electric field gradient (EFG) at the ^{19}F probe nucleus, the only stable isotope in F, by introducing the probe into samples. However, the ground state of ^{19}F does not have quadrupole (Q) moment because the nuclear spin of the state is $I = 1/2$, signifying that the ground state can not be applied for the

determination of EFGs. In the present study, therefore, we have focused our attention on the second excited state of ^{19}F ($I = 5/2$) fed through the β decay of ^{19}O to determine the controversial quadrupole moment of the state. 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 operations. Therefore, we are now working on the development of an online PAC measurement system coupled to an accelerator, where the recoil ^{19}O nuclei produced by the nuclear reaction of $^{18}\text{O}(d, p)^{19}\text{O}$ can be implanted kinetically in the sample of interest, so as to determine the Q moment by making use of a theoretical EFG.

P16 Study of dilute magnetism for Co and Mn codoped ZnO by means of emission Mössbauer spectroscopy

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Local structures and dilute magnetism in $\text{Zn}_{1-2x}\text{Co}_x\text{Mn}_x\text{O}$ (where $x = 0, 0.01, \text{ or } 0.03$) synthesized by a solid-phase reaction were investigated by means of X-ray diffraction (XRD) method and Mössbauer spectroscopy. It was found from the XRD patterns that all the samples have a hexagonal wurzite structure of ZnO matrix with no secondary phases detected. Local fields at ^{57}Co impurities were measured by emission Mössbauer spectroscopy, and no ferromagnetism was observed at room temperature. For the spectral analysis, therefore, we assumed two doublet components. The values of the isomer shift and quadrupole splitting of the main components observed for each sample showed good agreement with each other within their uncertainties. It was found

from their agreement with the literature values that Co^{2+} ions contributing to the main component occupy substitutional Zn^{2+} sites. As for the minor second component, it was observed that its fraction is raised with increasing concentration of the co-dopants, Co^{2+} and Mn^{2+} , in ZnO. For detailed understanding of local structures and dilute magnetism in ZnO, mere room-temperature measurement is not sufficient. We are thus now working on the introduction of a low-temperature Mössbauer spectrometer in our facility.

P17 ^{121}Sb Mössbauer spectra of pentacoordinated stiboranes with Au(I) fragments

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Rf_2SbAuL (Rf = mono anion of hexacumyl alcohol : L = $(\text{PPh}_3)_2$, $\text{P}(p\text{-Tol})_3$, $\text{P}(o\text{-Tol})_3$, PMes_3 , PCy_3 , PEt_3 , SMe_2) are synthesized to obtain information on the Sb–Au interactions. ^{121}Sb Mössbauer spectra of $\text{Rf}_2\text{SbAuP}(o\text{-Tol})_3$ (**3**), $\text{Rf}_2\text{SbAuPEt}_3$ (**6**) and $\text{Rf}_2\text{SbAuSMe}_2$ (**7**) are measured at 12 K using a $\text{Ca}^{121\text{m}}\text{SnO}_3$ source (16 MBq). The Mössbauer spectra are typical ones for trigonal bipyramidal stiboranes and have large negative quadrupole coupling constants (e^2qQ). Isomer shift (δ) and e^2qQ values for **3**, **6** and **7** ($\delta = 2.39 - 2.45$ and $e^2qQ = -22.1 - -23.9 \text{ mm s}^{-1}$) are intermediate between those of stiborane $[\text{Rf}_2\text{Sb}][\text{NET}_4^+]$ (**I** : $\delta = -1.37$, $e^2qQ = -24.9 \text{ mm s}^{-1}$) and a typical pentacoordinated stiborane $\text{Rf}_2\text{Sb}(p\text{-Tol})$ (**A** : $\delta = 4.16$, $e^2qQ = -18.3 \text{ mm s}^{-1}$). These results indicate the electron densities in the equatorial plane are greater than that of **A**. The same results are obtained for the similar hypervalent Sb compounds having the Sb–M (M = Fe, Ru, Cr, Mo, W) σ bond. Both

the δ and e^2qQ values for **3**, **6** and **7** are close to those of organoiron fragments such as $\text{FeCp}(\text{CO})_2$ and $\text{FeCp}(\text{CO})\text{PPh}_3$.

P18 Development of the measurement of neutron-induced reaction cross-sections of the minor actinides

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High-level radioactive waste which was produced by driving the nuclear reactor includes long-life nuclides, especially Minor Actinides (MA) such as Np, Am, and Cm. MAs are kept completely isolated from the biosphere, but their radiation may influence the environment for thousands of years to come. To solve that problem, it is suggested reprocessing MAs as fuel and transmuting MAs into other short-life nuclides by an Accelerator Driven reactor System (ADS). In the development of such systems, accurate and precision data of neutron-induced fission and capture cross-section of MAs are necessary.

The neutron-induced fission and capture cross-sections of ^{241}Am which is typical MA have been measured using various techniques. Our experiment was performed at the JAEA J-PARC facility. ^{241}Am sample is sealed up with an aluminum case. Therefore fission fragments cannot be measured directly. However, simultaneous measurement of prompt neutrons and gamma-rays by liquid scintillation counters allows to decide the cross-sections. In the

presentation, the neutron-induced fission and capture cross-sections obtained by this technique will be demonstrated and discussed the results in detail.

P19 Development of rapid dissolution and pre-concentration in sequential analysis of Pu and Am in soil samples

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Pu and Am isotopes generated in nuclear fuel are supposed to be released from Fukushima Nuclear Power Plant in March, 2011. In a sequential analysis at the trace level of Am and Pu, it is necessary to remove interfering substance through repeated purification and separation. In this study, we aim to establish a quantitative analysis of Pu and Am in a soil sample in a rapid manner of the processes. We focused attention on the improvement of the problems of dissolution and pre-concentration. First, we examined the dissolution by ammonium nitrate fusion, and the concentration by manganese di-oxide sorption. Next, we compared the filtration method with the electrodeposition method for the preparation of an americium α -ray source. After that we studied the application of these methods to soil samples from Fukushima.

P20 Trial production for gamma-ray emitting source applied by a 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 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.

P21 Investigation for Measurement of Strontium-90 by using Cherenkov Counting

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Strontium-90 (Sr-90) is one of the most important radionuclides among fission products because of its long half-life and its retention by the body.

Gas Proportional Counter (GPC) is often used for the Sr-90 measured in environmental samples, Cherenkov Counting by Liquid Scintillation Counter (LSC) has also been studied for a long time, it is known that described in the environmental radiation monitoring method.

In order to measure low-level concentration environmental samples accurately, it is necessary to evaluate background and efficiency, because FOM (figure-of-merit) for LCS is lower than GPC. The experiments were carried out to compare FOM with various vials by measuring the efficiency and the background. The result shows that low density poly-vial (inner Teflon coating) has high FOM and an advantage economically.

P22 Gamma-ray spectrometry of radionuclides in standard sample with detection efficiency curve using chemical reagents

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Detection efficiency of the HP-Ge spectrometer must be known in order to determine activities from the gamma-ray spectra observed. Detection efficiency is generally calculated with activities of radioactivity standard volume sources. The radioactivity standard volume sources contain several radioisotopes were only used in radiation controlled area. In the general laboratories, radioisotopes can't be used for measurement of environmental radioactivity. In this study, the detection efficiency curve was obtained by using KCl and LaF₃ as commercially available chemical reagent. Determined detection efficiency curve was validated by quantitative measurement of geochemical reference rock sample and radioactive standard sample. Reference value was in agreement with analytical results. The efficiency calibration using commercially available reagents will be useful outside radiation controlled area.

P23 Effect of pH on the Uptake and Retention of Tellurium and Cesium in Radish (*Raphanus sativus* var. *sativus*) Cultured under Hydroponic Condition

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A significant amount of Te-127m (1.1×10^{15} Bq) was released into the environment by the Fukushima nuclear power plant accident. Although the radioactive Te may cause internal radiation exposure in living beings at the early stage of the accident, little is known about the environmental behavior of tellurium including radioactive Te. In this study, the uptake and retention of Te and Cs was experimentally investigated in radish cultivar (*Raphanus sativus* var. *sativus*). The plants were grown in hydroponic media for about 30 days, exposed to media containing 1.0 ppm of Te and Cs for 2 hr, and continued to culture for 7 and 10 days. The concentrations of Te and Cs in the harvested taproot and leaves were quantified by ICP-MS. Since the chemical form of Te changes depending on pH, two types of solutions with different pH (3.2-3.8 and 6.3-6.8) were used. In the plants soaked to the solution of pH 3.2-3.8, the concentrations of Te and Cs varied largely among individual plants, and the relative standard deviation (RSD) was 4-117%. On the other hand, when the pH6.3-6.8 solution was used, the differences of Te and Cs concentrations among the individual plants were small, and the RSD was 6-17%. The concentrations of Te and Cs under neutral conditions were higher than those under acidic conditions. Therefore, it was suggested that variation of pH considerably effected uptake and retention of Te and Cs.

P24 The behavior of radioactive cesium in the northern and eastern region of Nagano prefecture

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The radioactivity measurement of various environmental samples (soil, water, leaf, plant, fish, and so on) in Nagano has been performed using hyperpure germanium detector to investigate the behavior of radioactive cesium from Fukushima Daiichi nuclear power plant. Radioactive materials from the reactors would migrate as radioactive aerosols, attaching to non-radioactive aerosols suspended in surface air. Thus, a potential radiation plume from the reactor site could reach to wide area of eastern and central Japan, including Nagano Prefecture; indeed, we first found several fission products on March 15, 2011. Since the aerosol particles attached radioactive materials efficiently deposit on the ground through the processes of wet depositions, the radioactive materials widely and variously distribute on the ground surface after wet depositions, migrating or being transported through various changes in environment. For the absorption into the body of a plant, radioactivity of ¹³⁴Cs included in various leaves collected in our university campus ranged between 1 and 10 Bq/kg, almost decreasing since 2011. For the deposition into soil, ¹³⁴Cs radioactivity in soils at Mt. Madarao ranged from 50 to 300 Bq/kg. There seems to be a correlation between ¹³⁴Cs concentration and altitude.

P25 Monitoring of radiocesium in Tamagawa River watershed after Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant Accident

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The Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident brought about radioactive contamination in the aqueous environment by ¹³⁴Cs (half-life: 2.06 y) and ¹³⁷Cs (half-life: 30.07 y) released from reactors. At present, activity concentrations of ¹³⁴Cs and ¹³⁷Cs migrated in environmental water are not significantly high. Trace-level activity concentrations in Tamagawa River watershed of Tokyo Metropolitan area have been determined to investigate behavior of Fukushima-derived ¹³⁴Cs and ¹³⁷Cs. The water samples and sediments were collected at 15 sampling locations in the Tamagawa River watershed. The concentrations of ¹³⁴Cs and ¹³⁷Cs dissolved in river water have been observed by gamma-ray spectrometry combined with the concentration method using ammonium molybdophosphate (AMP). The dissolved ¹³⁴Cs and ¹³⁷Cs were not detected in upstream within Okutama area. On the other hand, dissolved ¹³⁴Cs and ¹³⁷Cs in midstream and downstream in Tamagawa River watershed were detected.

P26 I-129 concentrations in soil samples collected at East Japan area -- Influence of the Fukushima Daiichi nuclear plant accident

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(Japan Chemical Analysis Center)

A large amount of radioactive materials were released after the Fukushima Daiichi nuclear plant accident in 2011, resulting in large-scale deposition of radionuclides onto the ground. In this study, we measured I-129 in surface soil samples collected in East Japan area to evaluate

the influence of the accident.

Soil samples were collected in 10 prefectures in 2010 and 2012, before and after the accident. Iodine was volatilized from the samples by using electric combustion-tube furnace and trapped into aqueous solution. Stable iodine (I-127) concentrations were determined by ICP-MS. After adding carrier, iodine in aqueous solution was purified through solvent extraction and converted into silver iodide for AMS (Accelerator Mass Spectrometer) measurement. I-129/I-127 ratios were determined by using AMS at JAEA Mutsu site. Finally, I-129 concentrations were derived from I-129/I-127 ratios and I-127 concentrations.

Increase of I-129 concentrations was found in the soil samples collected in 9 of 10 prefectures excluding Ibaraki. The ratios of I-129 concentrations after and before the accident ranged from 1.2 to 3.6. The atomic ratios of I-129/I-127 also increased. These results indicate the influence of I-129 released by the accident. In the soil sample of Ibaraki, conspicuous increase was not observed because of the relatively high background of I-129 due to a nuclear fuel reprocessing plant located in the region.

Further investigation will be needed to evaluate the distribution of I-129 in wider area in Japan.

P27 A trial of radioactivity measurement on radioactively-contaminated water including clay soils

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(Osaka Univ.)

In Japan, a large amount of radioactive materials has been released from the Fukushima Daiichi nuclear power plant in March, 2011. It

has been needed to analyze radioactive pollution of water and soils on paddy fields because of effective reduction of radioactivity in crops. In this research, we have tried to analyze radioactivity (^{137}Cs) of radioactively-contaminated water including clay soils.

Test sample water including soils was prepared through mixture of radioactively-contaminated soil into pure water. The sample was separated into coarse and fine particles and solvent with a sieve (sieve gauge: 125 μm) and a filter paper (diameter: 25 μm). Fine particles under 25 μm , which passed through the filter paper, were flocculated by the electrocoagulation method and then were collected with another filter paper. Radioactivity (^{137}Cs) of each classified component was measured by a face-type NaI(Tl) scintillation system constructed for low radioactivity measurement.

Measurement results showed that about 80% of the total-radioactivity (^{137}Cs) of the test sample came from the fine particles (under 125 μm). The particles under 25 μm were able to be filtered from the solvent by the electrocoagulation method. It was also confirmed that the radioactivity of the resultant solvent was about one order lower than the particles, which indicates that radioactive cesium in the soil is not easily soluble in water.

P28 Tritium concentrations in Atmospheric environment and rain at Fukushima Prefecture (2013~2014)

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After FDNPP accident, the tritium levels in

river water, pond water, and seawater in Fukushima Prefecture were investigated, the recent results show that those tritium concentrations (except the seawater close to FDNPP) are almost same as pre-accident level. But the data about atmospheric tritium are very few. We present the recent tritium levels of atmospheric HTO and rain at Fukushima Prefecture.

Atmospheric HTO and rain were continuously collected each month from Oct 2013 at Tamura City (Fukushima Pref.) and Fukuoka City (control site). The HTO collected on molecular sieve 3A (MS-3A) were desorbed by heating 400°C with flowing N₂ gas. The rain sample was enriched electrically. The sample mixed with scintillation cocktail (Ultima Gold uLLT, PerkinElmer) was measured 1000 min by low background liquid scintillation counter (LB-5, LB-7, Hitachi Aloka).

The atmospheric concentrations of HTO from Oct 2013 to April 2014 in Tamura city and Fukuoka city were below 1 Bq/L, and there were no significant difference between two sampling sites.

P29 Distribution of Artificial Radionuclides in Abandoned animals in the Ex-evacuation Zone of the Fukushima Daiichi Nuclear Power Plant Accident

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The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident released large amounts of radioactive substances into the environment. In order to provide basic information for biokinetics of radionuclides and for dose assessment of internal exposure brought by the FDNPP accident, we determined the activity concentration of radionuclides in the organs of 300 cattle, 50 pigs within a 20-km radius around the FDNPP. The radioactivity of the samples was determined by gamma-ray spectrometry using three HPGe detectors. In all the specimens examined, deposition of Cs-134 and Cs-137 was observed. Furthermore, organ-specific deposition of radionuclides with relatively short half-lives was detected, such as Ag-110m in the liver and Te-129m in the kidney. Regression analysis showed a linear correlation between the radiocesium activity concentration in whole peripheral blood (PB) and that in each organ. Thus, the activity concentration of Cs-134 and Cs-137 in an organ can be estimated from that in PB. The level of radioactive cesium in the organs of fetus and infants were higher than that of the corresponding maternal organ, respectively. Furthermore, radiocesium activity concentration in organs was found to be dependent on the feeding conditions and the geographic location of the cattle.

P30 Relationship between ¹³⁷Cs concentration in river water and inventory in catchment areas in eastern Fukushima Prefecture, Japan

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To investigate the factors controlling ^{137}Cs concentrations in river water, dissolved and particulate ^{137}Cs concentrations were measured at 16 sampling points in seven river systems under normal flow conditions during June–September in 2012 and 2013. The concentration of dissolved ^{137}Cs exhibited a strong correlation with the average ^{137}Cs inventory in the catchment area of each sampling point, whereas the concentration of particulate ^{137}Cs exhibited no clear relationship with the inventory. These results demonstrate that concentrations of dissolved ^{137}Cs in point samples of river water under normal flow conditions are determined primarily by the ^{137}Cs inventory of the catchment area upstream of the point.

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

P31 Migration pattern of riverine particles in the Agano River; implications from the FDNPP-derived low-level ^{134}Cs

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The FDNPP accident released large amounts of ^{134}Cs to the atmosphere and to land and sea surfaces over a wide region, particularly the Pacific Ocean-side of eastern Japan. In addition, marine sediments around the FDNPP were also subjected to radiocesium contamination by highly contaminated seawater as well as direct deposition on the sea surface and discharge of riverine materials. On the other hand, regardless of isolation from the ocean currents that pass the FDNPP and the small contribution by direct

input of radioactive depositions into the Sea of Japan, the FDNPP-derived ^{134}Cs was detected in estuary sediments as well as riverine particles in the Agano River which has the upper branch in Fukushima Prefecture and meets the Sea of Japan-side of central Honshu (Niigata Prefecture). In the present study, we examine the distribution of ^{134}Cs in the Agano River and the estuary area and discuss the transport mechanisms of the riverine particles.

P32 Penetration of actinoids and fission products in mortar and estimation of distribution using a partition equilibrium model

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Radionuclides on surface concrete would penetrate deeply by contact with precipitation and polluted water. The penetration is one of problem for decommissioning and demolition of the building of Fukushima Daiichi Nuclear Power Plant. Decontamination should be planned prior to works on the demolition. Scraping the contaminated part is required to decontaminate inside of the building. Radioactive strontium, uranium and plutonium, etc. are present in the polluted water in the building as well as radioactive cesium. In the present work, basic data of the penetration into the concrete were obtained to estimate the depth for decontamination.

Distribution coefficients, K_d , of Sr, Cs, U, Pu, and Am in a mortar sample were measured. In addition, ^{85}Sr , ^{137}Cs , and ^{238}U solution were passed through a column in which pulverized mortar was filled. Concentrations of the nuclides decrease exponentially with the depth. The

profiles in the mortar column were compared with the profiles estimated with a partition equilibrium model. The profiles based on the model agree well with the experimental profiles. Scraping a few millimeter would give enough decontamination for isotopes of Cs, U, Pu, and Am., while several centimeter of scraping would be required to remove Sr.

P33 Stability of DMPU and DMI for selective U(VI) precipitation against heating

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Cyclic urea compounds are potential precipitants with selectivity to U(VI) in acidic nitric acid solutions. Two candidate cyclic urea compounds, 1,3-dimethyl-3,4,5,6-tetrahydro-2(1H)-pyrimidinone (DMPU, 6-membered ring) and 1,3-dimethyl-2-imidazolidinone (DMI, 5-membered ring) were heated in HNO₃ to evaluate the quantitative stability. Heating was carried out for HNO₃ up to 4 mol·dm⁻³ (=M) containing 1 M DMPU at max. 70 °C up to 6 h. The residual ratio of the compounds was evaluated by the ¹H NMR spectra.

It was revealed that the degradation of DMPU was more enhanced with increasing temperature than with increasing concentration of HNO₃. The residual ratio of DMPU was found to be ca. 50% for the sample of 6 h heating in 4 M HNO₃ at 50 °C. For DMI, nearly no degradation was found to occur by heating for 6 h in 1 M HNO₃ at 50 °C. While, much more distinguished degradations were observed for the sample heated in higher concentration of HNO₃ and at higher temperature, resulting in the difficulty in

the evaluation of the residual ratios. An improvement in the determination of degraded DMI should be achieved for the evaluation.

P34 Temporal variations of carbon-14 concentrations in environmental samples in Ishikawa Prefecture

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The variation of ¹⁴C concentrations in atmospheric CO₂ samples collected every 10 days was measured in Dairahonmachi (36.5°N, 136.7°E) Kanazawa and Shikamachi (37.0°N, 136.8°E) Hakui, Ishikawa Prefecture, Japan in 2013. The variation of ¹⁴C concentrations in atmospheric CO₂ in Dairahonmachi kept constant from April to the middle of June, increased gradually to the early of September and decreased to December. The result in Shikamachi was similar to that of Dairahonmachi. We have already published the result that the long-term variation of ¹⁴C concentrations in atmospheric CO₂ was measured in Ohkuwamachi (36.6°N, 136.6°E), Kanazawa from 1991 to 2000. The trend of the variation of ¹⁴C concentrations in atmospheric CO₂ in Dairahonmachi was similar of that in Ohkuwamachi. The rice samples were collected in the same sampling points of atmospheric CO₂ samples in 2013. Compared the ¹⁴C concentrations in rice with the variation of those in atmospheric CO₂, the ¹⁴C concentrations in rice were closely the variation of those in atmospheric CO₂. It was found that the ¹⁴C concentrations in atmospheric CO₂ of summer had the influence on those in rice when rice was grown up. In other words, it's possible to use the ¹⁴C concentration of the rice as an index of the

^{14}C concentration in atmospheric CO_2 in the period while rice grows up.

P35 Variations of tritium concentrations and carbon-14 concentrations in environmental samples in Kanazawa city, Ishikawa prefecture after the accident at the Fukushima No. 1 nuclear power plant

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Environmental tritium decreased to levels lower than 1 Bq/L in 2000 in Kanazawa city (36.5°N, 136.7°E), Ishikawa Prefecture, Japan. A large amount of tritium was released from the Fukushima No.1 nuclear power plant caused by the damage of the nuclear reactors in consequence of the Great East Japan Earthquake on 11 March 2011. In this study, we measured the variation of tritium concentration in the precipitation of Kanazawa city after the day of the Fukushima No.1 nuclear disaster. One of the interesting result is that dramatic increases in the tritium concentration of 131.64Bq/L and 99.95Bq/L were observed on 30 May and 13 June in the variation pattern of the tritium concentration in the precipitation of Kanazawa city, although no marked change was observed in the period from April up to the end of May. This was done to estimate the influence of the tritium released into the environment by the Fukushima No.1 nuclear disaster on the tritium concentration in the precipitation of Kanazawa city. The variation of ^{14}C concentrations in atmospheric CO_2 samples collected every 10 days was measured in Kanazawa City from March to September in 2011. The effect by the Fukushima No.1 nuclear disaster was not find in the variation of ^{14}C concentrations in atmospheric CO_2 .

P36 Quality assurance of ^{129}I analysis by AMS

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The spent nuclear fuel reprocessing plant located in Rokkasho, Aomori, Japan (40°57'66"N, 141°21'87"E) is in going operation tests with actual spent nuclear fuels, and releases a limited amount of ^{129}I into the surrounding environment. We will determine ^{129}I concentrations by AMS in surrounding environment in Rokkasho Village, for evaluation of the effects of the release on the environmental ^{129}I concentration level. We confirmed ^{129}I analysis by certified reference materials.

This work was performed under contract with the Government of Aomori Prefecture.

P37 Establishment of Analytical Method for Technetium-99 in Soil

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Technetium-99(^{99}Tc) is one of the most important radionuclides for the environmental monitoring because of the long half-life and high fission yield. However, ^{99}Tc has not been determined in routine monitoring due to the low concentration and pure soft beta ray emitter. Therefore, an analytical method of ^{99}Tc was developed for environmental soil samples. In this research, the pretreatment procedures of soil were examined using both combustion and acid leaching methods.

At first, 10 g of soil sample in which ^{99}Tc (2 Bq) was added was used in recovery test of both methods, respectively. In the combustion method,

^{99}Tc was volatilized at 1000°C and trapped to alkaline solution in a combustion apparatus. In the leaching method, ^{99}Tc was leached with 3 M HCl for 2 hours. ^{99}Tc in these samples was purified with an extraction chromatographic resin (Eichrom, TEVA), and was measured by inductively coupled plasma mass spectrometer (ICP-MS). As a result, the recovery of ^{99}Tc by the combustion method was lower than 40%, but the recovery by the leaching method was approximately 80%.

Then, ^{99}Tc was added to 50 g of soil sample, and it was analyzed by the previous leaching method. As a result, the recovery was approximately 70%. So the leaching method could be applied to the ^{99}Tc measurement of the surface soil samples as a pretreatment procedure.

P38 A study for aging effect of I-127 and I-129 used sequential extraction

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Some of recent studies reported about the aging effect of iodine isotopes between stable iodine (^{127}I) and ^{129}I ($T_{1/2}=1.57\times 10^7\text{y}$): the former has been coming from the sea for the years of more than millennial scale, the latter has been generating from atmospheric nuclear testing, spent nuclear fuel reprocessing plants in Europe since 1960's. Some amount of ^{129}I was also released from the Fukushima Dai-ichi nuclear power plant accident at 2011.

To investigate the aging effect of the Fukushima accident derived ^{129}I in soil, it is important to know what kind of component the ^{129}I combines with. In this study a progressive

sequential extraction method was newly developed to obtain only the iodine sticking to the soil organic component. The stable iodine can be quantified by direct analysis of the fraction and ^{129}I can be quantified by Accelerator Mass Spectrometry (AMS) with the method of the fraction added with carrier. The fraction of the organic component for ^{127}I and ^{129}I can be evaluated respectively by comparing with the other fraction and/or with the total concentration obtained by the bulk analysis (e.g. by the pyrohydrolysis).

P39 $^{224}\text{Ra}/^{228}\text{Ra}$ ratios in coastal lagoon and the water dynamics in Lakes Nakaumi and Shinjiko

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In general, water dynamics in lagoon water are able to understand by examining the salinity of brackish water. However, the presumed water dynamics cannot present the time series changes of waters. Otherwise we are able to understand the water mixing processes of old and new waters, by applying radioactive elements, such as a short-life element as ^{224}Ra ($t_{1/2}=3.66\text{d}$) and a long-life ^{228}Ra ($t_{1/2}=5.75\text{y}$) in coastal lagoon.

We examined the $^{224}\text{Ra}/^{228}\text{Ra}$ ratios of both surface and deep waters from 20 locations in Lake Shinjiko. Both ^{224}Ra and ^{228}Ra in the water were collected by using a Mn-impregnated acrylic fiber. Measurement of ^{224}Ra was carried out within one and a half days by a Delayed Coincidence Counting system (RaDeCC) for α -decay and ^{228}Ra was measured by a γ spectrometry after one month of ^{224}Ra examination.

The results indicate the $^{224}\text{Ra}/^{228}\text{Ra}$ ratios in the surface water were 0.3-0.48 in western part

and 0.19-0.20 in eastern part of Lake Shinjiko. The deep water was generally lower than those of the surface, suggesting that the deep water is more stagnant and old in age. We judged Ra isotopes in water were derived from a shallow coastal area and were not from deep water.

P40 Migration pattern of the Tsushima Warm Current; Implication from $^{228}\text{Ra}/^{226}\text{Ra}$ ratio

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(¹LLRL, Kanazawa Univ., ²Nat. Res. Inst. Fish. Sci., ³Tokyo Univ. Agricul.)

The circulation of the surface water in the Sea of Japan is primarily governed by the lateral movements of the Tsushima Warm Current. Radium isotopes are continually supplied from the shallow shelf and coastal sediment in the continental-side of the East China Sea. Reflecting the short half-life of ^{228}Ra (5.75 y), within the Honshu Island-side of the Sea of Japan as well as the East China Sea, ^{228}Ra exhibits marked temporal and lateral variations, being caused by the seasonal change of the mixing ratio of ^{228}Ra -rich continental shelf water and the ^{228}Ra -depleted Kuroshio Current water. Therefore, ^{228}Ra is the strong tracer for the migration patterns of the Tsushima Warm Current. In the present study, employing low-background γ -spectrometry, we conducted observations in the lateral profiles of $^{228}\text{Ra}/^{226}\text{Ra}$ ratio on surface in the Sea of Japan in July of 2009–2014 and assess the migration patterns of the Tsushima Warm Current in summer.

P41 Selection of reagents for measurement

of low concentrations of ^{10}Be and ^{26}Al by AMS

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Cosmogenic nuclides of ^{10}Be and ^{26}Al are used especially for estimation of exposure age and erosion rate of rock. In order to measure traces of ^{10}Be and ^{26}Al by accelerator mass spectrometry (AMS) in these estimation, reagents with low levels of ^{10}Be and ^{26}Al for carrier and so on are required. In this study, ratios of $^{10}\text{Be}/^9\text{Be}$ or $^{26}\text{Al}/^{27}\text{Al}$ in several reagents were compared. The reagents were oxides for evaluation of background in measurement and standard solutions for atomic absorption spectrometry (AAS) or inductively coupled plasma optical emission / mass spectrometry (ICP) for carrier and evaluation of process blank. The ratios were measured in JAEA-AMS-TONO. The ratios of $^{10}\text{Be}/^9\text{Be}$ in the Be oxides and the Be standard solutions were approximately 10^{-15} - 10^{-14} levels. The Be standard solutions for AAS and ICP supplied by Merck had lower ratios of $^{10}\text{Be}/^9\text{Be}$ than those supplied by Wako and Kanto chemical. In this poster presentation, we will report the ratios of $^{26}\text{Al}/^{27}\text{Al}$ in several Al reagents.

P42 Possibility of radiocarbon dating to bronze implement

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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°C or more that CO₂ was extracted with 90% or more of yield. Next, the verdigris samples were extracted from the bronze implements of known archaeological age and the radiocarbon dating of them was performed. The calibrated radiocarbon ages included the corresponding archaeological ages. Considering a time lag from burying or rejection of bronze implement to generation of verdigris, archaeological age should be older than calibrated radiocarbon ages. Although, this result shows that the time lag can be neglected and that verdigris is a suitable sample for radiocarbon dating.

P43 Activity ratios of thorium and radium isotopes leached from Ishikawaite

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The leaching behavior of ²²⁴Ra, ²²⁶Ra, ²²⁸Ra, ²²⁸Th, ²³⁰Th and ²³²Th from Ishikawaite, radioactive mineral, was observed along pH of

HCl and HNO₃ aqueous solutions in contact. The activity ratios of ²²⁴Ra/²²⁸Ra and ²²⁸Th/²³²Th in the leachates from an Ishikawaite increased with an increase in pH of the HCl and HNO₃ aqueous solutions, while those of ²³⁰Th/²³²Th and ²²⁶Ra/²²⁸Ra decreased. The leaching behavior of ²²⁴Ra, which is formed through four decay processes of ²³²Th, was different from that of ²²⁸Ra, which is formed by a single decay of ²³²Th. ²²⁸Th, decay products of ²³²Th, was also leached more effectively than ²³²Th. The leaching efficiency of ²²⁴Ra and ²²⁸Th was estimated to be mainly dependent on the difference in the history of decay processes which have formed the nuclides. However, ²²⁶Ra and ²³⁰Th, which are decay products of ²³⁸U, showed different leaching behavior from ²²⁴Ra and ²²⁸Th. The difference in the leaching efficiency of ²²⁶Ra and ²³⁰Th may be attributed to the ratio of U/Th in the mineral.

P44 Variation of ⁷Be and ¹⁰Be concentrations in the atmosphere near the Earth's surface

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Concentrations of ⁷Be and ¹⁰Be in the atmosphere near the Earth's surface were measured at Tokyo during 2002 to 2013. Almost same seasonal variations were observed for 2002 to 2013: Be isotopes concentrations and isotopic ratios of ¹⁰Be/⁷Be were high during March to June by Stratosphere-Troposphere exchange (STE), and in other periods, concentrations were controlled by the vertical mixing in the troposphere. The annual average of ⁷Be and ¹⁰Be concentrations and the relative ⁷Be and ¹⁰Be

concentrations calculated from the neutron flux which observed in Delaware, were compared. These values become maximum at 2009, and the residence time of Be isotopes were estimated to about 2 years.

P45 Lateral profile of $^{228}\text{Th}/^{228}\text{Ra}$ ratio in seawater from the East China Sea and the Sea of Japan

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The East China Sea is an area of high productivity. Therefore, studies of material cycling have been conducted from the viewpoint of particle transport as well as water circulation. Activity of reactive ^{228}Th (half-life, 1.91 y) in seawater is predominantly governed by biogenic particle behavior, and therefore the combination of ^{228}Th and ^{228}Ra activities ($^{228}\text{Th}/^{228}\text{Ra}$ ratio) in seawater samples is useful for the study of scavenging processes. In the East China Sea, the $^{228}\text{Th}/^{228}\text{Ra}$ ratio of surface waters is low except for a wide variation in the Kuroshio-dominated area. On the other hand, the scavenging mechanism is not clarified in surface of the Sea of Japan. In the present study, using low-background gamma-spectrometry, we investigate the lateral and seasonal variations of $^{228}\text{Th}/^{228}\text{Ra}$ ratio in surface waters from the East China Sea and the Sea of Japan and assesses the scavenging mechanisms of ^{228}Th in these areas.

P46 Distribution of Cosmogenic ^7Be and ^{10}Be in the Northern North Pacific

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We collected seawater from the Northern North Pacific during R/V Hakuho-Maru KH12-4 cruise and investigated the distributions of ^7Be ($T_{1/2}=53.3$ day) and ^{10}Be ($1.36\times 10^6\text{y}$). The concentration of ^7Be and ^{10}Be in surface seawater were 20-110 and 150-1050 atoms/cm³, respectively. The concentration of ^7Be was high in the western north Pacific and low in the eastern north Pacific. And ^{10}Be was almost constant throughout all sampling point (except for the BD07 and BD15 station). Because of two stations lied high biological productivity, we considered deposition rate from the surface of Be was faster, and the concentration of the surface seawater had become low.

We showed vertical profile of ^7Be and ^{10}Be . The estimated ^7Be flux from the atmosphere to the sea surface were 0.013-0.048 atoms/cm²/sec. And the mean residence time of Be when used as a surface 10m were 0.13-0.80 year. There was a correlation between mean residence time of the Be and the maximum concentration of chlorophyll a.

P47 An optimization of magnetite dissolution method using oxalic acid for cosmogenic nuclide exposure age dating

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The cosmogenic nuclide exposure age dating by accelerator mass spectrometry (AMS) of the long half-life radionuclides produced in the rock

on the earth surface by secondary cosmic rays is used for geomorphological study etc. The determination of ^{10}Be and ^{26}Al in quartz is a main method for the cosmogenic nuclide exposure age dating. Here, we suggested the cosmogenic nuclide exposure age dating using magnetite as a new method. In the magnetite method, it is possible that five nuclides (^{10}Be , ^{26}Al , ^{36}Cl , ^{41}Ca , and ^{53}Mn) more than quartz method can be measured by AMS. In our previous study, concentrations of ^{10}Be and ^{36}Cl in magnetite separated from a basalt obtained at the Atacama Desert in Chili were measured. A cosmic-rays exposure age could be obtained from the ^{10}Be and ^{36}Cl concentrations and it was confirmed that the magnetite method is effective. However, improvement in initial magnetite dissolution was required due to long dissolution time and large solution volume. Therefore, in this study, magnetite dissolution conditions were optimized by dissolution examinations using commercial magnetite. The optimized solution, temperature, and time for 1-g magnetite are 15-mL aqueous solution of 5-g oxalic acid dihydrate, 70°C, and 6 days, respectively.

P48 Synthesis of endohedral 85-Sr metallofullerene by nuclear recoil using high energy neutron

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To obtain an endohedral metallofullerenes with high specific radioactivity using nuclear recoil phenomena, many experiments have been performed so far. However, production yield of metallofullerene by nuclear recoil using high LET radiation was found to be very small because radiation damages to targeted fullerene

molecule were not negligible. In order to increase the yield of radioactive metallofullerenes, it is very promising to employ the recoil of nuclear reaction by fast neutron from the viewpoint of reducing the radiation damage. In this session, we will report the synthesis of $^{85}\text{Sr}@C_{60}$ using nuclear recoil of ^{86}Sr ($n, 2n$) ^{85}Sr reaction by 14 MeV fast neutron.

Mixture of C_{60} with strontium salts such as $\text{Sr}(\text{NO}_3)_2$ and SrC_2O_4 were employed for the target materials. These samples were irradiated by fast neutron in Fusion Neutronics Source at Japan Atomic Energy Agency. After the irradiation, CS_2 , aniline, and HCl_{aq} were passed through the irradiated samples on the filter to dissolve C_{60} , $\text{Sr}@C_{60}$, and strontium salts, respectively. The gamma-ray from these solutions were measured by Ge detector. As a result, about 0.75% of radioactivity was observed in the aniline solution from SrC_2O_4 sample. This result strongly suggests that $\text{Sr}@C_{60}$ was efficiently produced by fast neutron irradiation of $\text{SrC}_2\text{O}_4/C_{60}$ target.

P49 Preparation of ^{109}Cd for fee-based distribution at the RIKEN AVF cyclotron

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(¹RIKEN Nishina Center, ²Japan Radioisotope Association)

Purified radioisotopes such as ^{65}Zn , ^{109}Cd , and ^{88}Y have been distributed to the general public in collaboration between RIKEN Nishina Center for Accelerator Based-Science and Japan Radioisotope Association since 2007. Cadmium-109 was produced by the $^{109}\text{Ag}(d,2n)^{109}\text{Cd}$ reaction by irradiating a Ag plate in a natural isotopic abundance with a

24-MeV deuteron beam from the RIKEN AVF cyclotron. So far, ^{109}Cd was chemically separated by filtrating the AgCl precipitates prepared after dissolving the irradiated Ag target. However, the chemical yield of this procedure was ~75%, not high enough, because of the coprecipitation of ^{109}Cd into AgCl. Therefore, we investigated the improvement of the chemical yield by chromatographic separation with HBr solutions by an anion exchange resin (Dowex 1X8, 100-200 mesh). The Ag was eluted with 7 M HBr from the column of anion resin and then ^{109}Cd was recovered with 3 M HNO_3 . The chemical yield was remarkably improved to be 98% by the newly applied procedure. The radioactivity of ^{109}Cd was determined by γ -ray spectrometry using a Ge detector. The radionuclidic purity was determined to be >99.9%. The chemical impurity and specific activity of the purified solution evaluated with ICP-MS will also be presented.

P50 Preparation of ^{85}Sr for fee-based distribution at the RIKEN AVF cyclotron

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Purified radioisotopes such as ^{65}Zn , ^{109}Cd , and ^{88}Y have been distributed to the general public in collaboration between RIKEN Nishina Center for Accelerator Based-Science and Japan Radioisotope Association since 2007. After the Fukushima Dai-ichi Nuclear Power Plant accident in 2011, discharged radio-strontium analyses are carried out and also some new extraction techniques are developed for analyses in various substances. Carrier-free

radio-strontium solution is useful in the evaluation for the performance of newly developed techniques. In this work, we investigated a novel procedure to prepare the purified ^{85}Sr solution having a high specific radioactivity using a no-carrier-added technique.

Strontium-85 was produced in the $^{nat}\text{Rb}(d,x)^{85}\text{Sr}$ reaction by irradiating a RbCl disk in a natural isotopic abundance with a 24-MeV deuteron beam from the RIKEN AVF cyclotron. Strontium-85 was chemically purified by the two-step chromatographic separation with a Sr resin (Eichrom, 100-150 mesh) and a cation exchange resin (Dowex 50WX8, 200-400 mesh). The radioactivity of ^{85}Sr was determined by γ -ray spectrometry using a Ge detector. The chemical impurity and specific activity of the purified solution were evaluated with ICP-MS for a control sample which was treated by the same chemical procedure as the irradiated sample.

In the present experimental conditions, the production yield of ^{85}Sr was about $3 \text{ MBq } \mu\text{A}^{-1} \text{ h}^{-1}$. The chemical yield of the purified ^{85}Sr solution was 89%. The radionuclidic purity was determined to be >99.9%. It was suggested that a high specific activity of $100 \text{ MBq } \mu\text{g}^{-1}$ or more can be achieved by the use of present chemical separation procedure.

P51 The substituent dependence on the yield of the unpaired electrons of soft X-ray irradiated pyrimidine DNA-bases

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In order to clarify the mechanism of DNA damage induced by K-shell ionization of

nitrogen and oxygen atoms, we measured short-lived '*in situ*' ESR spectra of pyrimidine DNA-base molecules at a synchrotron soft x-ray beamline BL23SU in SPring-8 (Hyogo, Japan) during the monochromatic soft X-ray irradiation. The ESR intensity for cytosine was significantly enhanced by the K-shell ionization, whereas that for thymine was not enhanced. These results suggest that cytosine favors to form the unpaired electron species. Further, by changing the substituent of the pyrimidine bases, we found out that the ESR intensities of pyrimidine bases were strongly correlated with the electronegativity of substituent, such as halogen atoms. The substituent effect of the pyrimidine ring in the DNA damage processes will be reported.

P52 Study toward total synthesis of ^{14}C -labeled C_{60}

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(Tokyo Metropolitan University)

Fullerene (C_{60}) is one of the most promising material in the organic chemistry, medical, and cosmetic field. For an application to the medical and cosmetic field, it is required to investigate detail pharmacokinetics of C_{60} because it is pointed out that some nanomaterials including fullerenes have nano-toxicity. The radiotracer method using C_{60} labeled with a radioactive carbon atom such as ^{14}C in its carbon framework is one of the most effective methods to study the pharmacokinetics of C_{60} . Production of ^{14}C -labeled C_{60} has been reported by Scrivens and coworkers using arc discharge method for the first time. Recently, we succeeded to produce ^{14}C -labeled C_{60} with the highest specific radioactivity to date by using a slightly improved Scrivens's method. However it is found that the increase in the production yield

and specific radioactivity by arc discharge method is extremely difficult. In addition, it is found that this method has some risks such as radioactive contamination by soot as a by-product of C_{60} .

In order to overcome these problems, we tried to synthesize ^{14}C -labeled C_{60} using total synthesis method because this method is possible to improve the yield and specific radioactivity of C_{60} without any radioactive soot.

P53 Production of medical radioisotope ^{90}Y with fast neutrons by the $\text{Be}(p,n)$ reaction

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The radioisotope (RI) for nuclear medicine is given an important role for a diagnosis with a high sensitivity and medical treatment for cancer today. In this study, we tried to optimize an effective production condition for ^{90}Y , which has been used for the nuclear medicine, by using a fast neutron provided in an accelerator. Particularly, in this experiment, we paid attention to a proton beam of 15-30 MeV provided from the small size cyclotron, which has been introduced into a medical institution for medical care such as the PET, coupled with a beryllium target, since the coupling has been reported a high energy neutron can be produced effectively. In this presentation we will report the excitation function of $^{90\text{m}}\text{Y}$ with the incident proton energy and evaluate the influence of by-products.

P54 Solvent extraction of astatine-211 for radiopharmaceutical use and its chemical form in the extraction process

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The ²¹¹At isotope with a half-life of 7.2 h has attracted attention as a promising α -emitter for radionuclide therapy. However, the chemical properties of astatine isotopes are not well known for application in radionuclide therapy. We focused on the DIPE-HCl extraction to study the chemical forms of astatine in the extraction process. The results will contribute to understanding the process of introducing ²¹¹At isotopes into the medicine.

We produced astatine isotopes by the reaction of ²⁰⁹Bi(α , 3n)²¹⁰At using the AVF cyclotron of RCNP in Osaka University. Astatine species were taken out from the irradiated targets through dry distillation and collected with various concentrations of HCl. The At species were extracted from the HCl solutions into equal volumes of diisopropylether (DIPE). The γ -activity of each phase in extraction was measured by a HPGe detector to obtain the distribution ratio of astatine isotope.

Dependence of the distribution ratio on the concentration of HCl was observed for DIPE. Distribution ratios are positively correlated with the HCl concentration more than 2 M, but are negatively correlated with that less than 2 M. Besides, chloride ion concentrations were found in the present study to make some effect on the DIPE-HCl extraction as well as those of hydrogen ion.