

English Abstracts

2S01 Application of nuclear probes and initiation of multitracer technique

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Radiochemical studies through my life are reviewed. Following hot-atom chemical studies by means of chemical separation in early days, chemical states of atoms produced by nuclear decays and reactions were studied by Mössbauer emission spectroscopy using short-lived source nuclides. For example, distribution of ^{119}Sn after EC decays of ^{119}Sb and $^{119\text{m}}\text{Te}$ in the lattices of SnSb and SnTe was determined. These results were further utilized to clarify the behavior of ^{119}Sb and $^{119\text{m}}\text{Te}$ recoiled by (p,2n) and (α ,2n) reactions in $^{120}\text{SnSb}$ and $^{117}\text{SnTe}$, respectively. Chemical states of these source nuclides, along with ^{57}Co , adsorbed on the surfaces of magnetic oxides were clarified through hyperfine magnetic interactions revealed on Mössbauer emission spectra.

Perturbed angular correlation of γ -rays was also utilized to study the chemical states of atoms after nuclear decay, ^{99}Ru in $\text{YBa}_2\text{Cu}_3\text{O}_{6.8}$, for example.

In the last decade at RIKEN, I mainly devoted myself to development of the multitracer technique, that is use of a number of radioactive nuclides produced by fragmentation of targets by heavy-ion reactions in order to trace many elements simultaneously. Chemical procedures to separate multitracers from different targets were established. All the tracers thus obtained in the solution are carrier-free. The multitracer solution contains about 50 elements in case of an Au target, for example. The multitracers have been applied to the uptake and distributions of various elements in plants and animals, to geochemical and environmental studies such as complexations by humic acids and to some basic chemistry of formation of metallofullerenes. More than 70 papers have so far been published on the application of multitracer technique.

2S02 Geo- and cosmochemical studies based on nuclear reactions in nature

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Isotopic variations caused by nuclear reactions in nature can be often used to decipher the past history of the geo- and cosmochemical materials and to understand their origins and formation processes. Mass spectrometry provides one of the accurate methods of measuring the stable and long-lived isotopes produced in the nuclear reactions. Recent developments of mass spectrometers make it possible to detect the isotopes of ultra-trace element with ICP-MS and observe the isotopes in sub-micro and nano meter region with SIMS. In this talk, I would like to present the following three topics on the isotopic characterization of planetary materials: (1) understanding of nuclear-chemical properties of the Oklo natural fission reactors and geochemical behaviors of fission products in geosphere, (2) finding of additional input of s-process isotopic component and initial abundance of presently extinct ^{135}Cs (half-life of 2.0×10^6 yrs) studied from Ba isotopic compositions of chondritic meteorites, and (3) geochemical determination of the double beta decay half-life of ^{100}Mo from Ru isotopic analyses of terrestrial molybdenites.

2S03 Spin crossover complexes- scientific significance of magnetic behavior and functions

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The multi functions of spin crossover iron(III) complexes are discussed after the basic properties of the spin crossover iron(III) complexes are introduced. The recent study of the spin crossover is focused on new functions produced by using spin crossover complexes of Metallomesogens, binuclear metal complexes and polymeric metal complexes because there have been over 70 years since the first report by Cambi and Szegö. The following topics are discussed in detail.

1. The difference of molecular structure between heat-excited electronic state and light excited one.
2. Metallomesogens with spin-transition phenomena
3. Reverse spin transition

We expect that practical materials are developed in near future.

2S04 Cosmic origins of life

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Origin of life on Earth has been contemplated largely with hypotheses counting on indigenous emergence or exogenous transfer of "life seeds" or panspermia. Comets and meteorites would serve as life-carrying "panspermian arks" to seed potentially habitable planets such as prebiotic Earth. Success of exogenous transfer depends on survivability of life encrusted in comets and meteorites, for example, as interplanetary and interstellar carriers. Likelihood of survival may be estimated on the basis of (i) hit rates and intensities of cosmic rays during transfer, and (ii) heat and mechanical shock generated by entry and landing impact. While the latter has been assessed despite uncertainties (velocity and angle of entry, presence of atmosphere/ocean, and size of a carrier), the former has been only conceptually considered. A major concern has been the hit of high-energy but low-frequency rays that react with comet/meteorite-building materials to generate secondary or sequential radiations, or hadron shower. The energy of hadron shower radiations may be high enough to damage life in shallow interior, and could be lowered to non-lethal levels in deep interior. Thus the occurrence of internal habitable zones, or protectable bodies, depends on the frequency and energy of hit rays and the size of life carriers.

3S01 Chemical studies of rutherfordium and nobelium on an atom-at-a-time scale

TOYOSHIMA, A.
(Advanced Sci. Res. Center, Japan Atomic Energy Agency)

Chemical studies of element 104, rutherfordium (Rf) and element 102, nobelium (No) in aqueous solution conducted at the JAEA tandem accelerator will be presented. Anion-exchange behavior of ^{261}Rf produced in the $^{248}\text{Cm}(^{18}\text{O}, 5n)$ reaction was systematically investigated in hydrofluoric acid and nitric acid mixed solutions using a rapid chemistry apparatus to elucidate fluoride complexation of Rf. It was found that Rf forms the hexafluoro complex $[\text{RfF}_6]^{2-}$ which is the same species as the lighter homologues Zr and Hf, and formation of the Rf complex is much weaker than that of the homologues. Oxidation of ^{255}No produced in the $^{248}\text{Cm}(^{12}\text{C}, 5n)$ reaction was studied using an electrochemistry apparatus combined with a chromatographic separation technique available for single atoms. It was substantiated that No^{2+} is successfully oxidized to No^{3+} in α -hydroxyisobutyric acid solution using the present apparatus.

1H01 Residual radioactivity at Atomic-Bomb dome
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(Grad. School Eng., Hiroshima Univ.)

Atomic-Bomb dome was constructed in 1914 as an products exhibition center. In 1996, A-Dome as well as Miyajima are registered as world heritages by UNESCO. Many persons are afraid whether residual radioactivity still exists in Hiroshima and affects people in there. It is clear that residual radioactivity is negligibly small today, however one can detect neutron-induced activities from iron samples, bricks and copper samples in Hiroshima by using high sensitive modern detectors. We have collected various samples from the A-Dome during past twice preservation construction. (1) In 1990, the arch shaped iron frame at the top of the A-Dome was exchanged to new frame. A-bomb exposed iron plates were sampled. Neutron induced ^{60}Co was observed from the irons samples.(2) In 2002-2003, during the third preservation construction, we have sampled copper plates from the A-Dome. The sample was cut for three pieces and dispensed to US group. (3) At that time, the ground was excavated to construct supports for preservation. Small copper pieces which were copper plates covered the Dome were found from the ground. Measurement of ^{63}Ni (half-life:100y) produced $^{63}\text{Cu}(n,p)^{63}\text{Ni}$ were performed to evaluate fast neutron fluence from the A-bomb.

1H02 Measurement of ^{63}Ni produced by the Hiroshima atomic bomb

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From the analysis of radionuclides produced by the thermal-neutron capture reaction, it was pointed out that the derived neutron spectrum for the Hiroshima atomic bomb is harder than the spectrum given in the Dosimetry System 1986 (DS86). In decommissioning the FM/FF cyclotron at Institute for Nuclear Study, University of Tokyo, we measured the residual radioactivities of ^{55}Fe , ^{60}Co and ^{63}Ni . During the process of these measurements, it was found that the ^{63}Ni produced by $^{63}\text{Cu}(n,p)^{63}\text{Ni}$ provides a unique measure to estimate the fast-neutron fluence of the Hiroshima atomic bomb. Thus, as basic nuclear data for the estimation, the excitation function for $^{63}\text{Cu}(n,p)^{63}\text{Ni}$ was measured using p-T, d-D and d-T neutrons from the 4.5 MV dynamitron accelerator at Fast Neutron Laboratory of Tohoku University, and the ^{63}Ni in copper samples exposed by the Hiroshima atomic bomb was determined. The chemical separation scheme for nickel previously developed in decommissioning the FM/FF cyclotron was improved. The ^{63}Ni separated from the copper samples was measured by liquid scintillation method. The results obtained for the Hiroshima samples are consistent with the values calculated by a new Dosimetry System 2002 (DS02).

1H03 Identification of plutonium released from Nagasaki atomic bomb

—Approach of measurement of $^{240}\text{Pu}/^{239}\text{Pu}$ isotope ratio by ICP-MS—

SAITO-KOKUBU, Y.

(JAEA)

Study of plutonium released from Nagasaki atomic bomb in the environment provides information for understanding of behavior and migration of plutonium for several decades, because the source of the release was evident and Nagasaki is one of the oldest cities where plutonium was deposited. Alpha-ray spectrometry of Pu was mainly used to assess the plutonium. However it was difficult to discriminate the

plutonium due to atomic explosion in Nagasaki from global fallout by $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio. In this study, $^{240}\text{Pu}/^{239}\text{Pu}$ ratio measured by ICP-MS was used to identify the source of Pu. Depth profile of the Pu in a sediment core collected at Nishiyama reservoir, Nagasaki city was determined to obtain depositional record of the Pu from the atomic bomb for 60 years. The profile revealed that the plutonium from the atomic bomb was deposited abruptly after the explosion and also deposited together with that from the nuclear tests. The Pu was measured in surface soils collected around the hypocenter of the atomic bomb to identify geographical distribution of the Pu from the atomic bomb. The results of the ratios indicated that the plutonium was deposited to about 100 km east from the hypocenter.

1H04 Dosimetry system for atomic bombs: DS02 and dosimetric calculation after DS02

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The first atomic bomb was dropped in Hiroshima on August 6, 1945, and the second atomic bomb was dropped in Nagasaki on that August 9. After these, high frequency incidences of cataract and leukemia were observed on the atomic-bomb survivors. Dose estimation was necessary to study a radiation effects on the atomic-bomb survivors. The dosimetric system were established as Tentative dosimetric system 1965 (T65D) in 1965 and Tentative dosimetric system (T57D), 1957. These were tentative systems and were expected for necessary modification in the future. It was stated the substantial atomic bomb dosimetric reevaluation in 1981 and established as Dosimetry System 1986 (DS86) in 1987. Amounts of neutron dose was reduced to 1/5-1/9 in DS86 from T65D.

To establish precise dosimetric system after DS86, it was started reevaluation from 2000. The new dosimetric system 2002 (DS02) was established in 2003. In this system, working group in the USA re-calculated source term and radiation transport with recent data. The DS02 is good accuracy for dose estimation, but It was not an essential change about the results from DS86. The out line of DS02 is introduced in this presentation.

After DS02, we made several check for DS02. Recently, we estimate dose of radiation from activated materials induced by atomic bomb neutrons. This is for a dose estimation of early entrance survivor. The “early entrance survivors” who were located far away from the hypocenter at the time of the atomic bomb explosion and entered the exposed area later. Possible exposure of the “early entrance survivors” is due to radiations from activated materials by atomic bomb neutrons. In this sense, activated materials are the main factor for “early entrance survivors”. Dose estimation of radiation from activated materials by atomic bomb neutrons is introduced as our recent calculation.

1H05 Assessment of individual radiation dose of atomic bomb survivors

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(Radiation Effects Research Foundation)

At ABCC (Atomic Bomb Casualty Commission) and RERF, a long-term epidemiologic study has been conducted for about 120,000 survivors. The estimated radiation risks derived from the study are unprecedented, while it became possible only because individual doses could be estimated. For this purpose,

various parameters were needed; e.g., distance from the hypocenter and shielding conditions including the type of houses, but such information was taken nearly 10 years after the exposure, which could be a source of uncertainty. The only possible indices for biological dosimetry are chromosome aberration frequency in blood lymphocytes and measurement of bicarbonate radicals in tooth enamel by means of electron spin resonance (ESR). Current summary will be presented.

1A01 Sequential Ion-Exchange Separation for Isotopic and Quantitative Analyses of Trace Amounts of U, Th, Pb and Lanthanoid

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(JAEA)

The authors investigated separation scheme of trace amounts of U, Th, Pb and lanthanoid using an anion-exchange column to analyse the abundance and isotopic ratio of those elements in environmental samples. An ICP-MS calibration standard containing U, Th, Pb, lanthanoid and the other elements of 50 ng each was used as a sample for this investigation. It was found that mixed-media of acetic acid, HCl and HNO₃ was good eluant for the separation of the interested elements, particularly, the separation of lanthanoid from Th. By using the mixed-media of 90% acetic acid and 0.5 M - HNO₃, alkali metal and alkaline earth elements, which are mostly major constituents of environmental samples, were washed out, but the interested elements were kept on the column bed. Lanthanoid was completely separated from Th by eluting with the mixed-media which consist of 80-90% acetic acid, 0.7-0.9 M HCl and more than 0.7 M HNO₃. Lanthanoid, Th, Pb and U were eluted sequentially with two types of the mixed-media of acetic acid, HCl and HNO₃; the mixture of methanol and HCl; 9M-HCl; 0.1M-HCl. This sequential ion-exchange separation technique will be applied to isotopic and quantitative analyses for environmental samples.

1A02 Quality control and uncertainty of measurement in ultra-trace analysis of nuclear materials

–Toward higher reliability in the safeguards environmental sample analysis–

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JAEA (Japan Atomic Energy Agency) has been conducting the environmental sample analysis, which is one of the IAEA's safeguards measures. In the analysis, isotope ratios of ultra-trace amounts of nuclear materials (uranium and plutonium) in samples taken from nuclear facilities are determined to detect 'environmental signature' of undeclared nuclear materials and activities. In order to keep the reliability of the measurement results, validation of analytical methods and estimation of uncertainty of measurement in such ultra-trace analysis should be properly done in accordance with worldwide standards. In this presentation, the authors report current activities and future perspectives of achievement of QA/QC (quality assurance & quality control) including estimation of uncertainty at CLEAR, a member of the IAEA's Network Analytical Laboratories for the environmental samples.

1A03 Decadal variation of ²⁴⁰Pu/²³⁹Pu atom ratios in water columns of the Japan Sea

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(Environ. Rad. Effects Res. Group, Natl. Inst. Radiol. Sci.,)

The atom ratio of ²⁴⁰Pu/²³⁹Pu is known to be a useful tracer to identify the sources of plutonium in the ocean. The objectives of this study are to measure the ²³⁹⁺²⁴⁰Pu activities and ²⁴⁰Pu/²³⁹Pu atom ratios in seawater from the Japan Sea and to discuss the transport processes of plutonium in the oceanic margin. The ²⁴⁰Pu/²³⁹Pu atom ratios were measured with SF-ICP-MS, which was equipped with a guard electrode to eliminate secondary discharge in the plasma and to enhance overall sensitivity. The ²³⁹⁺²⁴⁰Pu inventories were 42.8 Bq m⁻² obtained for 1984 and 86.6 Bq m⁻² obtained for 1993 in the Yamato basin. The inventories obtained for 1993 in the Yamato Basin were about two times higher than that (42.2 Bq m⁻²) of the expected cumulative deposition density of atmospheric global fallout at the latitude of 30 – 40°N. The atom ratios of ²⁴⁰Pu/²³⁹Pu showed no notable variation from surface to the bottom with an average ratio of 0.24. These high atom ratios proved the existence of close-in fallout plutonium originating from the Pacific Proving Grounds.

1A04 Rapid determination of short-lived radionuclides, ²⁴¹Pu and ²⁴¹Am in environmental samples using SF-ICP-MS

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(National Inst. Radiol. Sci.)¹

Studies on the distribution and behavior of ²⁴¹Pu and ²⁴¹Am in the environment is of great importance for radioecology and environmental radioactivity monitoring. Due to the decay of ²⁴¹Pu, ²⁴¹Am is continuously increasing and will reach its maximum activity in the middle of the 21st century. The activity ratio between ²⁴¹Pu and ²³⁹Pu is a useful tracer for source identification, similar to ²⁴⁰Pu/²³⁹Pu fingerprint, and the ²⁴¹Pu → ²⁴¹Am decay study could provide information about the release time of Pu in the environment. In addition, ²⁴¹Am is an ideal geochemical tracer to study the process of particle scavenging. Recently, we explored the feasibility of determination of ²⁴¹Pu and ²⁴¹Am in environmental samples using sector-field ICP-MS (SF-ICP-MS). We modified the anion-exchange chromatographic separation and purification procedure for Pu isotopes, with particular emphasis on reducing the background at m/z 241 as a result of elimination of PbCl⁺ polyatomic interference using HBr for Pu elution, instead of the commonly used NH₄I-HCl system. Using SF-ICP-MS with a high efficiency sample introduction system (APEX-Q), we achieved a detection limit of 0.5 fg/ml, or ca. 2 mBq/g. For ²⁴¹Am, we developed a rapid analytical method using a selective CaF₂ co-precipitation followed by extraction chromatography to effectively remove sample matrix and pre-concentrate ²⁴¹Am. Low detection limit of 0.32 fg/g or 0.041 mBq/g, which is better than that of alpha counting, was achieved. Applications to various environmental samples will be discussed.

1A05 Simplification of Tritium Measurement in the Environmental Water by means of Liquid Scintillation Counter

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Liquid scintillation counting is now the most popular method to measure the tritium concentration in the low level water samples such as environmental water samples. In the present environment water in Japan, the tritium concentration has become lower than 1 Bq/kg-H₂O which is within the

detective limit by the low background liquid scintillation counter. Then the electrolyte enrichment is needed before the liquid scintillation counting. It takes however much time with a lot of doing to achieve it. In the light of it, we have investigated to introduce new apparatuses and new techniques. As the result, we have achieved to measure at most 0.02 Bq/kg-H₂O with 2 days concentration and 1,000 min liquid scintillation counting. Using this new procedure, we have measured the tritium concentration of several sampling points for five years. Almost all of samples had the concentration between 0.1 Bq/kg-H₂O and 0.5 Bq/kg-H₂O.

1A06 An examination of simple purification method for automatic seawater tritium measurement

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(Japan Marine Science Foundation¹, National Institute for Fusion Science²)

The reverse osmosis membrane method as simple and quick desalination procedure was examined to application for seawater tritium measurement. The RO method is safe and saving electric power consumption, therefore, the method is suitable for automatic processing. Seawater samples artificial and sampling from harbor area and open ocean were purified by hand-operated desalinator (KATADYN Suvivor 06), and a part of desalinated samples was distilled by reduced pressure. The electro conductivity of the desalination samples was about 0.5 mS/cm, and the salinity of about 99% was removed from seawater samples salinity 33-34. There was no difference from desalination sample and distillation sample in the result of measuring by liquid scintillation counter after sufficient time from mixing with scintillator cocktail (Ultimagold-LLT). And decreasing tendency of chemical luminescence in immediately measuring after mixing with some scintillator cocktails (Ultimagold-LLT, Ultimafluo-M, Aquasol2) was also similar to desalination sample and distillation sample. In these results, the RO method can be applied as purification method for tritium measurement in the seawater sample.

1A07 Translocation of rhenium in a plant -Use of rhenium as a chemical analogue of technetium-

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(Biospheric assessment for waste disposal, Natl. Inst. Radiol. Sci.)

Technetium-99 (⁹⁹Tc) is of potential long-term importance in environmental dose assessment because it has a long half-life of 2.11×10^5 y and it is produced by thermal neutron fissions of uranium-235 and plutonium-239 giving a relatively high fission yield (ca. 6 %). The most stable chemical form of Tc in the surface environment is pertechnetate, TcO₄⁻, which has a high availability for plants. It is well known that Tc is highly accumulated in plants among non-nutrient elements, however, its retranslocate to other plant parts was not clear. In this study, therefore, a tracer experiment, growing plants (radish, *Raphanus sativus* L.) in a nutrient solution culture, was carried out to compare distributions of Tc and Re (as a chemical analogue of Tc) in plants, and then, Re translocation in a plant was studied. The results showed that the Re distribution pattern in a plant is the same as that of Tc, and both elements were highly concentrated in leaves through fine roots. The Re in leaves did not move to other plant parts, that is, Re was fixed in the leaves and no re-translocation was observed.

1A08 Behavior of environmental radioactivity in Mushrooms

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Although it has been more than 20 years from last nuclear weapon tests in the atmosphere or Chernobyl reactor accident, radioactive Cesium (Cs-137), which came from above events, is still detected in some foods. Especially, it is well known that mushroom shows the high concentration of Cs-137 compared with other foods.

In the present study, we investigated the behaviors of Cs-137 in various kind of wild mushrooms that were collected in the forest in Nanao city, Noto peninsula, Japan.

We report the difference in concentrations of Cs-137 between species, the uneven distribution of Cs-137 in the mushroom's body and the substratum-to-mushroom transfer factor (TF) of Cs-137.

In general, the higher concentration of Cs-137 was observed in the upper part of mushroom's body. The TFs of Cs-137 in the most of mushrooms were higher than those of K-40.

1B01 Development of Spent Fuel Reprocessing Process by Selective Sulfurization

- Study on the Pu Sulfurization by the use of ²³⁶Pu -

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For the recovery of fissile materials from spent nuclear fuel, a novel reprocessing process based on selective sulfurization was examined. This process consists of selective sulfurization of fission products (FPs) and dissolution of sulfurized FPs with dilute acid solution. In this study, the sulfurization behavior of uranium oxides and doped Pu by CS₂ was studied by using ²³⁶Pu tracer. The ²³⁶Pu tracer was produced using the reaction ²³⁷Np(γ, n)^{236m}Np → ²³⁶Pu. An encapsulated ²³⁷NpO₂ (²³⁷Np was 30 mg) in an evacuated quartz tube was placed behind a Pt converter, then, irradiated for 10 hours by bremsstrahlung from the electron LINAC of Laboratory of Nuclear Science, Tohoku University, where the average beam energy was 50 MeV. ²³⁶Pu tracer was isolated from ²³⁷Np and fission products. The isolated ²³⁶Pu was used for preparation of ²³⁶Pu doped U₃O₈ sample. This sample was sulfurized at 400 °C, 450 °C and 500 °C by flowing CS₂ gas for 1 hour. After sulfurization, the sample was immersed into 1M nitric acid solution and shaken for 1 hr at 50 °C. Then, the solution was separated by suction filtration to determine relative concentration of Pu and U in the solution by α spectrometry. The results indicate that uranium and doped plutonium are scarcely sulfurized and dissolved into 1M HNO₃ at sulfurization temperature below 500 °C, which is favorable for the separation of Pu and U from rare earth elements since those are supposed to be sulfurized and dissolved into 1M HNO₃ under this condition.

1B02 Adsorption behavior of f-elements on tertiary pyridine resin in aqueous alkali metal chloride solution

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The tertiary pyridine resin has two functions, one is a weakly basic anion exchanger and another is a soft donor ligand. These functions differently appear in different kinds of solutions or depending on the adsorbing ions. In the present work, the adsorption behavior of f-elements on the tertiary pyridine resin in aqueous alkali metal chloride solution is investigated by

using chromatographic technique. It was confirmed that actinides (An: Am and Cm) were eluted after elution of lanthanides (Ln), and Cm is eluted before Am. These elution tendencies are similar to the case of hydrochloric acid system. On the contrary, it was found that the lighter lanthanides is eluted ahead of the heavier lanthanides except Gd, and this elution order is quite opposite of the case of hydrochloric acid system.

1B03 Separation of Actinide ion based on Electrolytic extraction by Flow Electrolysis Cell for Rapid Ion Transfer
KITATSUJI, Y.¹, OKUGAKI, T.², KASUNO, M.³, MAEDA, K.², KIMURA, T.¹, YOSHIDA, Z.¹, KIHARA, S.⁴
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The novel solvent extraction and separation method for actinide ions based on the electrolytic ion-transfer at the interface between aqueous and organic solution phases was developed. Flow Electrolysis Cell for Rapid Ion Transfer (FECRIT) was employed to measure the voltammogram for the transfer of U, Np and Pu ions between aqueous and 1,2-dichloroethane phases. The cell was consisting of a porous Teflon tube in which Ag wire was inserted, a Pt wire placed outside of the tube and an electrolysis vessel. The tube and the Pt wire were set in organic solution in the vessel. Aqueous solution was forced to flow through the narrow gap between the Ag wire and the tube immersed with organic solution, so that relatively large area of aqueous/organic solution interface was formed at the surface of inside of the tube. Bis(diphenylphosphinyl)methane (BDPPM) or bis(diphenylphosphinyl)ethane (BDPPE) was employed as ionophore to facilitate the transfer of actinide ions, and transfer potential for each ions was determined. Based on the results of voltammetric studies, the separation of actinide ions such as U, Np and Pu is feasible by connecting the column electrode system prior to FECRIT to control the oxidation state of the ions.

1B04 Study on solvent extraction of Eu(III) and Am(III) using dithiocarbamate derivatives.
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Dithiocarbamate derivatives such as dibenzylammonium dibenzylthiocarbamate (DBzDTC), didecylammonium didecylthiocarbamate (DDDTC), dioctylammonium dioctylthiocarbamate (DODTC) and di-2-ethylhexylammonium di-2-ethylhexylthiocarbamate (EHDTC) were synthesized by the reactions of their secondary amines with carbon disulfide in diethyl ether. Using the synthesized dithiocarbamate derivatives, solvent extraction of ²⁴¹Am(III) and ^{152, 154}Eu(III) were carried out. Trivalent americium was well extracted by their extractants, but Eu(III) was poor. The distribution ratio (*D*) of Am(III) increased in order of DBzDTC > DDDTC ≈ DODTC > EHDTC, when the concentration of their dithiocarbamate derivatives was 0.050 mol/dm³. And values of *D*_{Am(III)} and *D*_{Eu(III)} decreased with an increase of the hydration-ion concentration of aqueous solution. The slope of log *D* vs log[H⁺] was three. Further, the values of *D*_{Am(III)} and *D*_{Eu(III)} increased with an increase in the concentration of DBzDTC in organic phase. The slope of log *D* vs log [DBzDTC]_{org} was also three. These results indicate that

three dithiocarbamate derivatives are required in order to extract Am(III) and Eu(III) into organic phase.

1B05 Measurement of stability constant of lanthanide, americium, curium, and californium in HIBA/acetate system using capillary electrophoresis and its application to short-lived nuclides produced with an accelerator

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For understanding of the effects of difference between 4f- and 5f-orbital on chemical bond, we have systematically investigated the chemical behavior of lanthanide and actinide, which include those only produced in the nuclear reactions. In this study, the values of electrophoretic mobility of lanthanide, americium, curium, and californium were measured in HIBA/acetate system using a capillary electrophoresis technique. The order of their values agrees with that of the ionic radii for tripositive ions with coordination number of larger than 8, regardless of whether the ions are lanthanide or actinide. Therefore, the ionic radius of Cm³⁺ was estimated 108.5 ± 20 pm by comparing the mobility of curium with those of lanthanide tracers. We also attempted to apply this capillary electrophoresis technique to short-lived nuclides produced with an accelerator.

1B06 A relation between mutual separation behavior and structural parameters in lanthanide 2-hydroxybutyrate complexes

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It is important to obtain structural parameters such as bond distances and bond angles to study about chemical properties of heavy actinide (An) and lanthanide (Ln) ions. In this study, structures of lanthanide (except Promethium) and Yttrium containing 2-hydroxyisobutyrate (HIB⁸) were identified by X-ray analysis. The complexes were prepared by the reaction of each lanthanide ion with excess amount of 2-hydroxyisobutyrate in aqueous acetic acid at pH 2-3. X-ray analysis shows that the one-dimensional polymer [Ln(HIB)₃(H₂O)] (Ln = Ce-Tb) and the discrete molecular complex H[Ln(HIB)₄] (Tb-Lu) were afforded. The coordination number for all of the complexes is eight and sum of the bond distances decrease with increasing the atomic number to give a line against ionic radius of the lanthanide at C.N. = 8. It is found that the separation behavior of the lanthanide and sum of the bond distances has a linear correlation.

1B07 Preparation and crystal structure of U(VI) coordination compound containing 2,2'-bpy ligand

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Actinoid chemistry is highly related to the reprocessing of nuclear fuels and treatment of actinoid wastes in the backend chemistry of today's operating nuclear power plants. The fundamental investigation of the bonding the structure and uranium coordination compounds gives important information in the field of backend chemistry. A new U(VI) coordination compound (2,2'-bpy)H[UO₂(acac)(NO₃)₂]•(2,2'-bpy) has been prepared. Crystal structure of the new

(2,2'-bpy)H[UO₂(acac)(NO₃)₂]•(2,2'-bpy) has been determined using a suitable single crystal. 2, 2-bpy molecule is not coordinated to U(VI) atom.

1B08 Structural Determination of Neptunium Species in Aqueous Solutions by EXAFS and Quantum Chemical Calculations

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Neptunium (⁹³Np) is one of the most problematic nuclides in the nuclear fuel reprocessing process and the following radioactive waste disposal because of its chemical similarity to the fissile (*i.e.* reusable) nuclides of uranium (U) and plutonium (Pu). Proper understanding of the Np behavior in the reprocessing process or in the migration process on the geological disposal of radioactive wastes requires vast fundamental information about the chemical properties of Np in solutions. As a matter of fact, extensive investigations have been carried out for the last several decades to reveal the chemical properties of Np in solution. However, despite a large number of precedent works, the complex structure of Np species in solution is still not understood sufficiently. In the present study, Np solution samples with different oxidation states are electrochemically prepared in various aqueous solutions, and the complex structure of Np species in the sample solutions are determined by EXAFS spectroscopy, partly with the help of DFT calculations.

1B09 Instrumental neutron activation analysis of cosmetics

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Many cosmetics claiming inclusion of Pt, Au and Ag are available through the Internet, one hundred yen shop and specialty cosmetic shops. The manufactures have to indicate the ingredients of cosmetics and are not allowed to add the forbidden elements into cosmetics by the regulation. However, these cosmetics don't have labels of indicating the percentage of the ingredients. So, we analyzed these cosmetics by using instrumental neutron activation analysis (INAA), and studied qualitatively by a scanning electron microscope-electron probe X-ray microanalysis (SEM-EDX). Twelve kinds of samples and 5 kinds of standard samples were analyzed. The samples were irradiated in JRR-3 reactor (JAEA) for 10 min at a neutron flux of $5.2 \times 10^{13} \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$. One sample visually included gold colored grains, the grains were found to contain gold by the SEM-EDX. The size of the gold grains was from 10 μm to some mm. One sample, which claims inclusion of nanometer-size Pt, was analyzed by the SEM-EDX and the Pt grains sizes were 100-200 nm. For INAA, cosmetics samples on filter papers could be irradiated after drying up under an infrared lamp. The lower detection limit of Pt of the standard sample was 1 ppm under the present experimental condition of INAA. Some samples contained Pt, Au or Ag as claimed in the label by the producer, however, some samples' Pt were less than detection limit contrary to the claiming in the label. The percentage of the Pt ranged from 1 to 80 ppm among the samples.

1B10 Neutron activation analysis of Hf in Zr metal using an internal standard method

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Neutron activation analysis with an internal standard method was applied to the quantification of Hf in high purity zirconium metal. Zr was used as an internal standard to compensate for in-homogeneity of the neutron flux in an irradiation capsule, improve the repeatability of gamma ray measurements and linearity of the calibration curves of Hf. The effect of internal standard to reduce the uncertainties and the comparing the analytical results to the analytical results by ICP-QMS will be discussed.

1B11 Measurement of agricultural samples using MPGA

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Many of agricultural samples contain large amounts of hydrogen. Hydrogen emits a 2.2MeV prompt gamma ray in a neutron capture reaction. The low-energy photons due to the Compton scattering of high energy γ-rays from hydrogen obviously will cause an increase in the background in the lower region of the γ-spectrum, resulting in a decrease in the detection limit.

Hydrogen emits only one prompt γ-ray at a time at every neutron capture reaction. The multiple γ-ray detecting method, which is known as a coincidence method, is widely used in nuclear spectroscopy. In this method, only elements which simultaneously emit two or more prompt gamma ray at a capture reaction can be measured. Therefore, by applying the multiple γ-ray detection method to PGA, the influence from hydrogen can be reduced.

MPGA beamline was sealed by silicone rubber. The air in the beamline was replaced by helium or carbon dioxide in order to reduce the background from the air. Standard agricultural samples and polymer samples were measured by MPGA detector system. PGA and MGPA spectrum was compared. Advantages of using MPGA include the ability to identify and qualify.

This research was partly conducted as the Practical Application Research and supported by JST Innovation Satellite Ibaraki.

1B12 Completion of Multiple Prompt Gamma-ray Apparatus

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To achieve the new trace element analysis method of Multiple Prompt Gamma-ray Analysis (MPGA) we have completed the construction of the MPGA apparatus, STELLA, at C2-3-2 beam line at the neutron guide hall in JAEA Research Reactor, JRR-3.

In December 2007, an MPGA system made of eight clover Ge detectors has been completed, based on NEDO fund. A fast data acquisition system for this system has been developed and

achieved a comparable energy resolution and much faster speed of 6 micro second per event, which is more than ten times better than the old system. An automatic liquid nitrogen supply system for Ge detectors and an automatic sample changer enable the automatic measurement. Furthermore, in FY2007 we have started another project of Practical Application Research supported by JST Innovation Satellite Ibaraki and added two coaxial type detectors; rapid gamma-ray energy calibration and on-line quantification software is developed as well as necessary gamma-ray database for them.

In July 2008 we have started the application of STELLA to MPGA based on the Interorganization Atomic Energy Research program by University of Tokyo, Toray Research Center and JAEA. In the talk we present the performance and status of STELLA as well as those of the apparatus at the new facility of J-PARC MLF.

This research was partly conducted as the Practical Application Research and supported by JST Innovation Satellite Ibaraki. (The authors) would like to thank Director Katsutoshi Goto and staff for their assistance.

1B13 Measurement of standard samples by multiple prompt gamma-ray analysis

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(JST Innovation Satellite Ibaraki, JST,¹ Innovative Nuclear Science Research Group, JAEA,² Inorganic Analysis Laboratories, TRC³)

Neutron-induced prompt gamma-ray analysis (PGA) is valuable as a non-destructive multi-elemental analysis. In order to improve the sensitivity and resolution for the quantification of trace elements, we have developed Multiple Prompt Gamma-ray Analysis (MPGA). MPGA is a combination system of PGA and multiple gamma-ray detection method. In MPGA, two prompt gamma-rays emitted simultaneously from each of the radionuclides in a sample are simultaneously measured with a multiple gamma-ray detector assembly to construct a two-dimensional matrix and the individual nuclides are measured in a completely separate form without performing any artificial operation such as chemical separation. Furthermore, owing to the negligibly low residual radioactivity, the methods are suitable for analyzing of scarce samples.

We performed measurements of elemental standard samples by MPGA. As an example of the two-dimensional matrix, the matrix for the reference sample BCR680 is displayed in Fig.1. Based on the results, we present the characteristic and advantage of MPGA. The Ge detector system for MPGA was constructed at the neutron guide-hall of JRR-3M reactor in JAEA.

This research was partly conducted as the Practical Application Research and supported by JST Innovation Satellite Ibaraki. We would like to thank Director Katsutoshi Goto and staff for their assistance.

1B14 Analysis of cosmochemical samples by multiple prompt gamma-ray activation analysis

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We examined multiple prompt gamma-ray activation analysis (MPGA) apparatus, which was almost completely

constructed in the experimental hall of JRR-3 in 2007, to apply it for analyzing cosmochemical samples. The sensitivity of single measurement using MPGA apparatus was about 50 times larger than one of normal PGA apparatus in JRR-3. Several elemental content ratios in JB-1 and Allende meteorite were tried to be determined by an internal standard method. The determined Fe/Si and Ti/Si ratios in JB-1 were almost consistent with recommended values, but for Allende meteorite the determination values were not so good. Some studies such as a calculation way of obtaining a net count rate and influence of a rate of chance coincidence are required to get accurate determination values. In addition to the examination, strongest coincidence gamma ray pairs released from some elements were also found by irradiation of some chemical reagents.

1B15 Neutron Activation Analysis of Marine Samples by Short-life

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Analyzing trace elements in marine samples has difficulty because of high concentrations of Na, Cl, Mg, Br, etc. Also acid digestion is hard to complete because of the tolerance of high concentrations of polysaccharides in them. For these reasons, activation analysis was chosen to analysis several elements in marine samples in this paper. Several elements in sea mustards on different growing stages, cultivated oysters, and sea squid were analyzed by neutron activation. Sea mustards and sea squirt were collected from Onagawa Bay, Ishinomaki, Miyagi. Cultivated oysters were collected from several bays of Miyagi Pref. and Korea. Samples were washed with tap water, then distilled water, and freeze dried after the separation of each organ. After pulverizing the dried samples, 0.3 - 0.5 g of powders were sealed in plastic vials, and irradiated for 5 - 60 seconds in SLOWPOKE-2 Reactor in Dalhousie Univ., Canada. After 12 sec - 3min cooling time, gamma counting was done by Compton suppression system. Six elements, Ag, Al, Br, I, Se, and V, were determined by the activities of Ag-110 (half life 24.6 s, 657.76 keV), Al-28 (2.24 min, 1778.99 keV), Br-80 (17.68 min, 616.3 keV), I-128 (24.99 min, 442.9 keV), Se-77m (17.45 s, 161.93 keV), and V-52 (3.75 min, 1434.08 keV), respectively.

1B16 Aragonite-calcite phase transition of pelagic surface sediment dated by excess ²³⁰Th

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Aragonite, which originated in biogenic matters, has been alternated calcite by authigenic effect in deep-sea sediment. As compared with calcite, crystal structure of aragonite is due to be unstable slightly. However, the detailed *in situ* process of calcite-aragonite phase transition in sediments, especially the reaction time required for transition has not been clarified. In order to elucidate the crystal structure transition of calcium carbonate by high time resolution, XAFS measurements of calcium in pelagic sediment with detailed sedimentation age were performed in this study. All carbonates for measurement were low Mg calcite judging from the results of elemental and XRD analyses; therefore, crystal structures of carbonate in this sediment were calcite or aragonite. Ca K-edge XANES spectra of pelagic sediments of various sedimentation ages were

measured. The feature of spectrum of sediment whose sedimentation age was 1.6kyr had a strong resemblance to that of aragonite. Observed shoulder on K-edge around 4.04keV was small, and at higher energy region than K-edge, spectrum had the same feature as that of aragonite. Spectrum of 4.1kyr sediment was composed of that of aragonite and calcite by 1:1 ratio, and spectrum of 128kyr sediment had the almost same feature as that of calcite. The sequential spectra variation resulted from authigenic effect of aragonite-calcite phase transition, and crystal structure transition from aragonite to calcite in high time resolution was clarified by this measurement.

1B17 High-sensitive measurement of uranium L_{III}-edge X-ray absorption near-edge structure (XANES) for the determination of the oxidation states of uranium in crustal materials

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Uranium L_{III}-edge XANES spectra for natural rocks at a concentration range from 0.96 to 124 mg kg⁻¹ were measured using a log spiral bent crystal Laue analyzer (BCLA) combined with a multi-element Ge detector at SPring-8 BL37XU. The XRF spectra with and without BCLA indicated that U fluorescence is selectively extracted by using BCLA. It was found that the quality of the XANES spectra using BCLA is greatly improved due to reduction of interfering fluorescence from major components such as Rb and Sr. The ratio of signal to background intensities in the U L_{III}-edge XANES spectra increased by a factor of 2.9-17 with the use of BCLA, which greatly lowered the detection limit of U for XANES for the speciation of uranium in natural samples. In addition, it was demonstrated that the fluorescence XANES method coupled with BCLA enables the speciation of uranium for various natural samples such as acidic igneous rocks, ferromanganese nodules, sediments, and some sedimentary rocks such as shale and limestone.

1B18 Design of Quartz Fusion Furnace for Extraction of Carbon-14

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Cosmogenic radionuclides are produced in rocks depending on the thick of surface ground covering them, which derives that erosion rate can be evaluated from the relation of accumulation and half life of cosmogenic radionuclide. Among rocks, quartz is chemically stable and abundant in the environment and hence it is useful material for evaluation of erosion rate and dating.

In the previous work, we had obtained CO₂ gas through combustion of graphite with a quartz powder sample collected from deep underground where no cosmogenic C-14 was produced and hence the isotopic ratio of the CO₂ gas was expected to agree with that of the graphite. The analysis yielded that a carbon recovery of 2 %, a C-14 date of 28,277 yBP and a delta C-13 of -5 per mil.

In this work, the analysis of the graphite, however, yields that a C-14 date of more than 46,000 yBP and a delta C-13 of -15.1 per mil. These values are quite different from previous

results. We assume that this large difference would be caused by the low carbon recovery rate and then investigate the effect of additive into quartz fusion furnace to increase recovery rate. As a result, the addition of CuO provides higher recovery rates and constant isotopic ratios.

1B19 Gas Experiment at Fermilab anti-proton target station

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Various radiations, such as proton, neutron, pion, γ -ray, and X-ray produced by bombarding high energy beam to a target, are present in a target station used for high energy nuclear physics during machine operation. We could assume high energy accelerator environment as a mimic environment of the upper atmosphere. We investigate transportation behavior of the nuclides using Fermilab anti-proton target station. 120 GeV-proton beam was introduced to the Inconel 600 target at Fermilab anti-proton target station. Various radionuclides which originate from the target and air are present in the target vault. Particulate and gaseous component were taken to the filter stack, ADVANTEC 5A filter, Teflon membrane filter with pore size of 0.1 μ m, charcoal filter, and NaCO₃-impregnated filter, using an air pump. After sampling, gamma-spectrometry was carried out. Approximately 70% of nuclides, produced by activation of air, were collected to the membrane filter, and 5% of other spallogenic nuclides, originate from target, were collected to the 5A filter. Halogen was observed in only charcoal filter. No radionuclide was detected in the NaCO₃-impregnated filter. It was confirmed that the radionuclides except for halogen migrate together with particulate material and the halogen migrates as gaseous state.

1P01 Change of concentrations of trace elements and proteins in hepatocyte of mice at early stage of zinc deficiency

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Concentrations of trace elements and proteins in hepatocyte of Zn-deficient and control mice were investigated. Eight-week old male mice of ICR strain were divided into two groups; one was fed with Zn-deficient diet, and the other with control diet. After one week of dividing, their livers were removed and separated into six subcellular fractions by centrifugal method. Concentrations of ten elements in each fraction were determined by instrumental neutron activation analysis. Furthermore, proteins in cytosolic fraction were separated into ninety fractions by gel-filtration chromatography. Then BCA protein assay was carried out for each fraction. For zinc concentrations in all subcellular fractions, there were no significant differences between two groups. On the other

hand, cobalt concentrations in several fractions of Zn-deficient mice were higher than those of control mice. No significant differences were found for the protein concentrations in each fraction separated by gel-filtration chromatography.

1P02 Measurement of Iridium using Neutron Activation Analysis with gamma-gamma Coincidence

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The multiple γ -ray detection method developed for nuclide quantification has achieved better resolution and sensitivity than the ordinary singles gamma-ray detection method. The INAA combined with this method is called NAAMG (Neutron Activation Analysis with Multiple Gamma ray detection)^{1,2)} and has been successfully applied to various fields. In the case of neutron activation analysis, measurements of γ -rays from trace elements are strongly interfered by the γ -rays from isotopes produced from major elements, e.g., ²⁴Na, ⁵⁶Mn. Therefore chemical separation processes are sometimes required to eliminate the major elements for determination of the trace elements. Measurements of iridium in geological sample were carried out by this method. Samples were irradiated at the JRR-3. Iridium concentrations in several standard rock samples and geological samples were also measured by this method. Fifty to one hundred mg of each sample were sealed in quartz tube and irradiated together with iridium standard. After irradiation, the γ - γ coincidence of multiple γ -rays from the Ir-192 was measured with the Ge detectors array, GEMINI-II. Gamma rays from about 10 ppt of iridium in geological samples can be detected by this method.

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1P03 Investigation of relation between transition of tideland and vertical distribution of elements by neutron activation analysis

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The adjacent areas of Yatsu tideland were reclaimed from 1960's to 1970's, so Yatsu tideland was isolated from Tokyo Bay. After that, a viaduct divided Yatsu tideland into two areas. We collected sediments vertically in a smaller area of two divided areas in order to investigate relation between transition of the tideland and vertical distribution of elements. Using instrumental neutron activation analysis and prompt gamma-ray analysis, we got vertical distribution of over thirty elements. Chemical states of iron were investigated by Moessbauer spectroscopy. Results of measurement clarified that vertical distribution of elements changed radically at ca. 20 cm and ca. 40 cm from the surface. According to concentrations of sulfur and chemical states of iron, sediments became anaerobic at ca. 40 cm from the surface. This is regarded as influence of reclamation. It is thought that the flow of seawater was weakened by reclamation, and supply of oxygen was reduced. On the other hand, the sediments became relatively aerobic at ca. 20 cm. This seems to be influence of the construction of viaduct. The separated area became like a waterway, so it is supposed that dissolved oxygen

concentration in water increased, and then became slightly oxie.

1P04 Study on the separation of Am(III) from Eu(III) using self assembled monolayers (2)

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The adsorption behavior of ^{152,154}Eu(III) and ²⁴¹Am(III) onto self assembled monolayers, which are (a) 3-mercaptopropyl functional silica gel (SAMs(1)) and (b) a reaction product of SAMs(1) with CS₂ (SAMs(2)), was investigated. It was proved that the adsorption of ^{152,154}Eu(III) and ²⁴¹Am(III) onto both SAMs depends on the pH value in the aqueous phase. When the solution contained 1.0 mol/dm³ sodium nitrate, it was shown that both SAMs had a higher affinity for ^{152,154}Eu(III) than for ²⁴¹Am(III).

1P05 Plutonium records in sediment core from Nagasaki Bay, Japan

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In this study, depth profiles of ²³⁹⁺²⁴⁰Pu concentration and ²⁴⁰Pu/²³⁹Pu ratio in the cores collected from the center of the Nagasaki Bay were determined to find depositional records of plutonium released from Nagasaki atomic bomb and nuclear tests. Sediments above 1.2 m were analyzed. The ²³⁹⁺²⁴⁰Pu concentrations were from below detection limit to 3.5 Bq/kg. The ²⁴⁰Pu/²³⁹Pu ratios ranged between 0.05-0.27. Plutonium was not detected below 1 m. This showed that sediments above 1 m were deposited after the explosion of the Nagasaki atomic bomb and World War II. Deposition of plutonium released from the Nagasaki atomic bomb will be also reported in the presentation. The concentration and the ratio in sediments at the depth above 1 m increased and their maximums were detected at about 0.5 m. These results indicated that the profiles were corresponding to depositional records of plutonium from the nuclear tests carried out around the Marshall Islands and the atmospheric nuclear tests.

1P06 Determination of ³⁶Cl in soil samples

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We report an improved sample preparation method for measuring ³⁶Cl in soil by accelerator mass spectrometry (AMS). Surface soils contain a lot of organic sulfur, which often leads to the isobaric interference from ³⁶S in ³⁶Cl-AMS. The leaching procedure was examined with several ways of Cl extraction, including different timing and amount of H₂O₂ addition to the extractant. The amount of residual sulfur in each sample was analyzed by ³⁶S counting at final detector per ³⁵Cl beam-current at ion source. The results showed that the addition of H₂O₂ to the extracted solution effectively reduced ³⁶S interference. We also developed a BaCO₃ precipitation method which removes SO₄²⁻ ions in the sample solution as a co-precipitation of BaCO₃. Tracer experiments of ³⁵SO₄²⁻ demonstrated that the

BaCO₃ precipitation method was effectively, removing more than 95% of SO₄²⁻ ions in the solution.

1P07 ^{239,240}Pu in the water columns of the Eastern Pacific
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The ocean, by virtue of its large surface area, has received a major share of the fallout Pu. With the aim of understanding the sinking behavior of Pu in water column, depth distributions of ^{239,240}Pu, ²³⁸Pu were measured in water columns of the Eastern Pacific. It was found that ^{239,240}Pu concentration increased from surface to 600 m and showed low concentration from 1000 m to the bottom. ²³⁸Pu was clearly detected in the southern hemisphere of the Eastern Pacific. ²³⁸Pu in the southern hemisphere is attributable to the SNAP-9A accident in 1964.

1P08 Distribution of ²⁴¹Am concentration in water column of the eastern Indian Ocean and its adjacent sea in 1997 and eastern Pacific in 2003.

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Artificial radionuclide ²⁴¹Am in the environment is the daughter of ²⁴¹Pu from atmospheric weapons testing. In this work ²⁴¹Am concentrations were determined in large-volume seawater samples collected at eastern Indian Ocean and its adjacent sea and eastern Pacific. The vertical distributions of the nuclides were compared with each other for a better understanding of the behaviour of the nuclides in the marine environment. In the closed Sulu Sea, the depth profile showed that ²⁴¹Am is sinking faster than ^{239,240}Pu. At a station in Indian Ocean, it is considered that the flow of great ocean conveyor belt affects the depth profile of ^{239,240}Pu.

1P09 The concentrations of natural radionuclides in groundwater samples after the Noto Peninsula Earthquake 2007 (a follow-up)

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A ruinous earthquake (M 6.9) struck Noto Peninsula on 25 March 2007. For more than 1 year after the earthquake, groundwater was sampled every month near the earthquake center to analyze ²³⁸U, ²³⁴U, ²³²Th, ²³⁰Th, ²²⁶Ra, ²¹⁰Pb and ²¹⁰Po. The results are as follows: (1) As for the Th isotopes, they were rarely detected in both places. (2) As for U isotopes, at Monzen station, ²³⁸U and ²³⁴U concentrations were high after the earthquake and tended to decrease with time and ²³⁴U/²³⁸U activity ratio was higher than 1. At Sugihira station, ²³⁸U and ²³⁴U concentrations increased gradually with time, but ²³⁴U/²³⁸U activity ratio was approximately 1. (3) As for Ra, the ²²⁶Ra concentrations were about 1 mBq/L at both places. (4) As for Pb and Po, ²¹⁰Pb concentrations decreased gradually with time and much higher than that of U and Ra at both places. ²¹⁰Po concentrations were lower than ²¹⁰Pb concentrations. These results suggest that the flow pass of groundwater was not changed largely by the earthquake, but a considerable amount of ²²²Rn dissolved in ground water system.

1P10 Characterization of Pu isotopes in soils of Gansu in northwestern China

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We investigated the total ²³⁹⁺²⁴⁰Pu activities and ²⁴⁰Pu/²³⁹Pu atom ratios in surface soils (0-5 cm) in the Kumtag Desert in western Gansu Province, and in a soil core sample in Lanzhou using sector-field ICP-MS. In the surface soils, ²³⁹⁺²⁴⁰Pu activities in fine particles (<150 μm) were 1.3 to 2.1 times of those in coarse particles which ranged from 0.005 to 0.157 mBq/g. ²⁴⁰Pu/²³⁹Pu atom ratios in the surface soils ranged from 0.168 to 0.192 with a mean of 0.182 ± 0.008. Surprisingly, this mean was like that of a typical global fallout value although the Kumtag Desert was believed to have received close-in fallout derived from Chinese nuclear tests mainly conducted in the 1970s. A mean ²⁴⁰Pu/²³⁹Pu atom ratio that was like that of a typical global fallout value was also observed in the soil core sample in Lanzhou. In the soil core sample, ²³⁹⁺²⁴⁰Pu activities in the various layers ranged from 0.012 to 0.23 mBq/g, and the inventory of ²³⁹⁺²⁴⁰Pu (32.4 Bq/m², 0 -23 cm) was slightly lower than that expected from atmospheric global fallout (42 Bq/m²) at the same latitude. Rapid downward migration of Pu isotopes was observed in Lanzhou soil core sample layers. The contribution of top 10 cm layer of surface soils to total inventory was only 17 %, while the contribution of deeper layers (10-23 cm) was as high as 83 %. The ²³⁹⁺²⁴⁰Pu activity levels and ²⁴⁰Pu/²³⁹Pu atom ratios in soils in Gansu Province, China are similar to those in atmospheric deposition samples collected in the spring season in recent years in Japan; this finding supports the hypothesis that the temporal change of ²³⁹⁺²⁴⁰Pu deposition in eastern Asia is controlled by the long-range transport of suspended soil dust particles originating from the East Asian arid and desert areas.

1P11 The problem and measure on the determination of Pb-210 and Po-210 in groundwater sample

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The Noto-hanto earthquake occurred on March 25th in 2007. It fact was informed that the radio activity on the ground was raised up extraordinarily at the center area of the earthquake place. Therefore, we started to pick up two groundwater samples in the area, and to determine ²¹⁰Pb and ²¹⁰Po in water every month. The ferric-hydroxide coprecipitation method was applied to preconcentration of the water sample. But several problems were occurred, for example escaping of lead from ferric-hydroxide precipitation and the sorption of polonium to the inside of a sample container and so on. So, we decided to clarify the condition of the coprecipitation about the preconcentration of each nuclide. As the matters which effected to precipitate separation, the species of carrier precipitate, the precipitate formation and dissolution, and the chemical form were examined. Also, we reviewed a measure from the blank activity of carrier reagent, and from the viewpoint of the processing of waste liquid.

1P12 Variation in Environmental Radioactivity in Water at Higashi-Hiroshima, Japan

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We have measured environmental radiations of water samples from the junction point to public sewerage system and Kadowaki-Chosetsu Pond in Higashi-Hiroshima Campus, Hiroshima University for 15 years. Seasonal dependence of the environmental radiation for the junction to public sewerage system was observed. The seasonal dependence was closely related to the weight of residue of the sample. In the present study, we picked up 10 water samples from Higashi-Hiroshima city. The environmental radiations and pH were measured monthly. The results were discussed from the point of microbial activity.

IP13 Secular variation of $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio in the summit products from Izu-Oshima volcano, Japan

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Observations were carried out on the radioactive disequilibria among ^{238}U - ^{230}Th - ^{226}Ra in the summit products of the post-caldera eruptions (9th C, 1421 AD, 1552 AD, 1684 AD, 1777-1778 AD, 1951 AD and 1986 AD) from Izu-Oshima volcano. Uranium and Th isotopes in the products were determined by isotope dilution method coupled with alpha-ray spectrometry. Radium-226 was determined by gamma-ray spectrometry. Activity ratios of $^{238}\text{U}/^{230}\text{Th}$ and $^{226}\text{Ra}/^{230}\text{Th}$ in the products were larger than unity, being enriched in ^{238}U and ^{226}Ra relative to ^{230}Th , which was often observed for fresh volcanic products from island-arc volcanoes. The concentrations of Th in the products tended to increase from 9th C towards 1777-1778 AD, and to decrease towards 1986 AD. The decay-corrected $^{226}\text{Ra}/^{230}\text{Th}$ activity ratios in the products between 9th C and 1777-1778 AD were relatively constant, and then tended to increase from the 1951 AD towards 1986 AD. These variations of the Th concentrations and $^{226}\text{Ra}/^{230}\text{Th}$ activity ratios in the summit products suggested that a new magma was recently supplied to the magma chamber beneath Izu-Oshima volcano.

IP14 Chemical states of Ca adsorbed on soil in connection with acid neutralization capacity

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Exchangeable cations play an important role on acid neutralization capacity (ANC) of soil. We have already reported that Ca had the biggest influence on ANC. But Ca of various forms is contained in soil and it is not clearly shown of which form Ca has affected ANC. So in this research we examined how Ca adsorbed by iron in allophane affected ANC of soil. Allophane including iron was synthesized according to Osaka's method. The allophane was stirred for 1 h in 0.01 mol/L calcium hydroxide solution to adsorb Ca ion. ^{57}Fe Moessbauer spectra were taken in order to know chemical states of iron. It was found the amount of structural iron increased and that of surface iron decreased after the reaction. This result showed that iron adsorbed Ca as inner-sphere complex. Next, the allophane was stirred for 1 day in pH 4.5 nitric acid to observe the role of iron on ANC. We took ^{57}Fe Moessbauer spectra and observed the reaction toward the opposite direction. So it turned out that the iron contained in allophane affected ANC of soil by adsorbing or releasing Ca. Moreover, extended X-ray absorption fine structure (EXAFS) spectra on the Ca K edge revealed that the length of Fe-Ca

bond was about 2.5 Å by analysis for 2nd shell of RSD spectra.

IP15 Distributions of ^3H , ^7Be , ^{22}Na , ^{137}Cs and Ra isotopes in rain water, ground water and river water of the Tedor River Basin

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Cosmogenic ^3H , ^7Be , ^{22}Na , global fallout ^{137}Cs and natural radium isotopes have a wide range of half-lives, various origins, source materials and geochemical behaviors, and are expected to be powerful tracers for water flow system. However, with the exception of radium isotopes and a peculiar case of ^{137}Cs in a contaminated aquifer, these nuclides have not been studied due to measurement difficulties using standard γ -spectrometry. In order to examine the distribution of these nuclides in inland waters of the Tedor River Basin, Ishikawa Prefecture, rain water, ground water, river water and coastal water samples were collected. A simple batch method using Powder ion-exchange resin was suitable for rapid treatment of a large volume (300 L) of ground water even on the sampling sites without sacrificing yields. Low-background γ -spectrometry combined with this chemical treatment enabled the determination of extremely low-levels of radionuclides in inland water samples. The measurement of these nuclides in water samples provide new informations on the flow system of inland water, including residence time, the mixing of precipitation, and aquifer's environment in this area.

IP16 Estimation of exposure age from measurement of ^{26}Al and ^{10}Be in Abukuma Penplain

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Cosmogenic ^{26}Al and ^{10}Be produced by cosmic ray spallation in quartz were measured by accelerator mass spectrometry (AMS) after chemical separation and purification. Stable aluminum concentration was measured by the Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES). Preliminary result of these radionuclides concentrations were obtained. The depth profiles of $^{26}\text{Al}/^{10}\text{Be}$ ratio could suggest the formed process and the post-changes of the river terrace, which was formed by deposition of the weathered granitic soil, in northern Abukuma Penplain, Japan. Also, the extraction method of cosmogenic ^{14}C from quartz has been developing for the ^{14}C dating of rock.

IP17 Cross section measurements for monoenergetic neutron-induced reaction at RCNP

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Cosmogenic nuclides are used as a probe to investigate the history of cosmic ray irradiation for extraterrestrial and terrestrial materials. Most cosmogenic nuclides are produced

by neutron-induced reactions. For these studies, accurate cross section data for neutron-induced reactions are strongly desired. However, in high-energy regimes (> 100 MeV), cross sections are poorly known for most elements. In this study, a high-energy neutron irradiation was performed to obtain cross sections for geochemically and cosmochemically important elements by using ${}^7\text{Li}(p, n)$ reaction at N0 beam line in RCNP, Osaka University. We carried out neutron bombardments with two experimental configurations; 0° and 30° from primary proton beam direction. The neutron spectrum of 0° consists of high-energy monoenergetic and low-energy continuum neutron components, and the neutron spectrum of 30° contains only the latter component. By subtracting the 30° spectrum from the 0° spectrum, quasi-monoenergetic neutron fluence is available. The radionuclides produced in these two neutron irradiation were analyzed quantitatively by a gamma-ray spectrometry and accelerator mass spectrometry (AMS). The production rates for neutron-induced reactions were obtained, and cross sections for monoenergetic 362 MeV neutron-induced nuclear reaction were determined.

1P18 Anion-exchange experiments of nuclear reaction products transported by He/KCl and He/KF gas-jet systems

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Radioactive nuclides of Nb and Ta were produced in the ${}^{\text{nat}}\text{Zr}/{}^{\text{nat}}\text{Hf}(p, xn)\text{Nb}/\text{Ta}$ and ${}^{\text{nat}}\text{Ge}/{}^{\text{nat}}\text{Gd}({}^{19}\text{F}, xn)\text{Nb}/\text{Ta}$ reactions at the JAEA tandem accelerator. The reaction products were rapidly transported by He/KCl and He/KF gas-jet transportation systems to the chemistry laboratory. It was found that the transport efficiency using the He/KF gas-jet system is comparable with that by the He/KCl gas-jet system which has been generally used in the chemical studies on the superheavy elements. Subsequently, on-line anion-exchange experiments with ${}^{88,90,90\text{m}}\text{Nb}$ and ${}^{170,178}\text{Ta}$ were performed in HF/HNO₃ solutions. A small physical adsorption of Nb on the surfaces of the resin and a column was observed when the products were transported by the KCl aerosols, while better reproducibility of the elution of Nb was obtained using the KF aerosols. We interpret that stable fluoro complexes of Nb are rapidly formed by using the KF aerosols and we can suggest that the He/KF gas-jet system should be used in the on-line anion-exchange experiment of the group 5 elements including Db for the investigation of the fluoride complexation of these elements.

1P19 Ion-exchange behavior of Zr and Hf as homologues of Rf in sulfuric acid solution

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In the present study, ion-exchange behavior of Zr and Hf in 0.018 – 0.991 M sulfuric acid was studied by a batch method to find out appropriate experimental conditions for an on-line experiment of Rf. Cation- and anion-exchange resin, MCI GEL CK08Y and CA08Y, were used. The results showed that the carrier-free radiotracers ${}^{88}\text{Zr}$ and ${}^{175}\text{Hf}$ in sulfuric acid were strongly adsorbed on the cation-exchange resin as well as the

anion-exchange one, indicating that cationic and anionic species coexist in sulfuric acid media. Ion-exchange behavior of Zr and Hf was very similar to each other and distribution coefficients (K_{ds}) of these elements decreased with increasing sulfuric acid concentration. Adsorption sequence on the anion-exchange resin was $\text{Zr} > \text{Hf}$, while that on the cation-exchange resin was $\text{Zr} < \text{Hf}$, confirming that Zr has a stronger ability to form sulfate complexes than Hf. Chromatographic separation between Zr and Hf can be expected in 0.028 – 0.991 M H₂SO₄ on CA08Y and 0.087 – 0.991 M H₂SO₄ on CK08Y in the on-line experiment.

1P20 New apparatus for aqueous chemistry of the heaviest elements

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Aqueous chemistry of element 105, dubnium (Db), is now being performed at JAEA. The nuclide 34-s ${}^{262}\text{Db}$ is produced in the ${}^{248}\text{Cm}({}^{19}\text{F}, 5n)$ reaction with the cross section of 1.5 nb. This results in the production rate of about 0.25 atoms per min. In order to carry out chemical experiments on Db with single atoms, we need to develop a new device to shorten the time for the preparation of α sources. A new apparatus based on a continuous sample preparation and detection system has been developed. For the HPLC system, we modified the ARCA. There are two pairs of sampling and detection systems, each has two sample transport arms on which thin Ta sheets (30 cm long) are placed. The effluent is continuously collected on this moving Ta sheet and evaporated. The dried sample is successively going into the α -particle detection chamber where an array of 12 PIN-detectors is installed for each arm. The sample preparation is accomplished within 20 s. Further, from the measurement of radioactivities in each detector, we can measure an elution curve of a nuclide for each condition. The new system enables us to perform cyclic column chromatographic separations of short-lived ${}^{262}\text{Db}$ nuclides and automated detection of α -particles.

1P21 Study on reversed phase chromatography with the TTA extractant for chemistry of Element 104, rutherfordium (Rf)

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Application of extraction with TTA is interesting in research on the solution chemistry of Element 104, rutherfordium (Rf), because it can determine the extraction species. In this work, we prepared resin containing TTA extractant (TTA-Resin) for reversed phase chromatography, and investigated solvent extraction behavior of its homologues, Zr and Hf using the TTA-resin to confirm whether it can be applied for Rf experiment. The batch extraction experiment was performed to measure the distribution ratios (D) by using the tracers of ${}^{88}\text{Zr}$ and ${}^{175}\text{Hf}$. Besides conventional liquid-liquid extraction was performed to confirm the extraction species with TTA. We also checked the equilibrium time of extraction process using the resin, which will be important for application to the Rf experiment. Result of the batch extraction shows that the D values of Zr and Hf fall suddenly due to formation of Zr or Hf complex with halide ions if the concentration of $[\text{F}^-]_{\text{eq}}$ or $[\text{HCl}]$ comes to be over a certain value. From the conventional

liquid-liquid extraction experiment, the TTA extraction species were found to be $M(TTA)_4$. The experiment for equilibrium time of extraction process is under way.

1P22 Cosmic ray BKG reduction system for Ge detector utilizing thin plastic scintillators and wavelength shifting fibers

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In order to reduce the contribution of cosmic ray components on background of Ge detector by anti-coincidence system, five thin plastic scintillators (PSs) (180 mm x 200 mm, and 3 mm in thickness; KURARAY SCSN-81) which covered 2π geometry connected with five wavelength shifting fibers (1 mm in diameter; BICRON BCF-91A) that was coupled to one thin PMT (1/2"; HAMAMATSU H3165-10) was examined. The PSs system was inserted into the passive shield of Ge detector. All events were recorded in list mode. Relative efficiency of PSs, time spectra, time dependences of coincidence events, multiple coincidence events, and coincidence and anti-coincidence spectra were tested. Counting rate of PSs, Ge, and coincidence events were roughly 1k, 130 and 50 min^{-1} , respectively. More than 2 PSs events a Ge event has sometimes been detected. This thin PSs system was sufficiently thick to detect cosmic ray components and more than 50% of those induced background reduction will be expected.

1P23 Incident Energy Effect of the Production Yield of Endohedral ^{133}Xe -Fullerene

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Incident energy effect on the production yield of endohedral ^{133}Xe -fullerene by ion implantation was investigated. Fullerene targets for ion implantation were made by vacuum evaporation of C_{60} on Ni foils. Implantation of $^{133}\text{Xe}^+$ ions into the targets was carried out with an isotope separator at acceleration energies of 30 keV. For retardation of incident energy, the high voltage of 15kV was supplied from a retardation power supply to the fullerene targets. After ion implantation, the fullerene part on the target was dissolved in *o*-dichlorobenzene. The solution was analyzed by high performance liquid chromatography (HPLC) for determination of endohedral ^{133}Xe -fullerene. The yield of endohedral ^{133}Xe -fullerene was defined as a percentage of the radioactivity of endohedral ^{133}Xe -fullerene to that of ^{133}Xe implanted into the target. The yield of endohedral ^{133}Xe -fullerene for 15 keV was about 30% larger than that of 30 keV. Those energy dependences of the production yields can be ascribed to the amorphization of endohedral ^{133}Xe -fullerene. The yields of the endohedral ^{133}Xe -fullerene should be more increased by decreasing incident energy.

1P24 Detection of muonic atom formed by muon transfer process

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Negative muon captured in matter result in the formation of muonic atom. Because its Bohr radius is roughly 200 times smaller than for an electron due to its much larger mass, it screens nuclear charge. Especially, muonic hydrogen atom has neutron-like properties because of the strong nuclear screening

by muon. Therefore, a muonic hydrogen atom can penetrate an electron cloud easily and transfer the muon to the deeper atomic levels of another atom. Such a capture process is called transfer capture. Other formation process for muonic atom is direct capture. In this research, we try to obtain information on the initial state of captured muon. We measured the muonic Ar X-ray emitted after the formation of muonic atom in order to reveal initial state of captured muon. The measurements were performed at RIKEN-RAL muon facility. Argon gas sample was irradiated to investigate direct capture process. $\text{H}_2+\text{Ar}(0.1\%)$, $\text{H}_2+\text{Ar}(0.3\%)$ mixture samples were measured to investigate transfer process. We analyzed μAr X-ray intensity of argon gas and mixture gas. Obtained intensity value of each transition was normalized by 2-1 intensity value. "2-1" represents muonic transition from principle quantum number $n=2$ to $n=1$. The measured relative intensities of muonic 3-2 and 4-2 X-rays for argon are much larger than one for mixture. We compared the experimental value of relative muonic X-ray intensities with the calculated one and discuss initial state of captured muon.

1P25 State analysis of iron in iron oxide-silica glass nanocomposites

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State analysis was carried out on iron in iron oxide-silica glass nanocomposites, manufactured by sol-gel processing. Starting iron compounds were FeSO_2 , $\text{Fe}_2(\text{SO}_4)_3$ and $\text{Fe}(\text{NO}_3)_3$. Sintering temperatures were tested from 700 to 1000 degrees Celsius. Samples prepared were then subjected to Mössbauer spectroscopy at room temperature and 4K and to X-ray diffraction. Magnetization was measured with a superconducting quantum interference device (SQUID). Among iron oxides, epsilon phase was dominantly formed in the samples prepared from iron sulfates, FeSO_2 and $\text{Fe}_2(\text{SO}_4)_3$, whereas alpha phase was dominantly formed in that prepared from nitrate, $\text{Fe}(\text{NO}_3)_3$. Gamma phase was found in all the samples tested, nevertheless its particle size was so small that it showed superparamagnetism. For practical applications such as magnetic recording, epsilon phase has the most propitious property; we then conclude that the iron oxide-silica glass nanocomposite prepared from iron (II) sulfate and sintered at 1000 degrees Celsius is the most favorable.

1P26 ^{170}Yb Mössbauer spectra of ytterbium(III) oxide and chloride

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Ytterbium-170 Mössbauer spectrum of ytterbium(III) oxide was measured at 20 K using a $^{170}\text{Tm}/\text{Al}_2\text{Tm}$ prepared by a neutron irradiation in JRR-4 T_B pipe of Japan Atomic Energy Agency. The spectrum clearly shows the existence of two different ytterbium sites assigned to C₃ symmetry and C₂ symmetry site of Yb_2O_3 . The spectrum is compared to that of gadolinium(III) oxide measured by ^{155}Gd Mössbauer spectroscopy.

1P27 ^{57}Fe and ^{155}Gd Mossbauer spectroscopic studies of cyano-bridged Fe(III)-Gd(III) complexes

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We have been prepared a new cyano-bridged Fe(III)-Gd(III) hetero-one-dimensional coordination polymer compound $\text{mer}[\text{Fe}(\text{CN})_3(\mu\text{-CN})_3\text{Gd}(\text{H}_2\text{O})(\text{o-phen})_2] \cdot 3\text{H}_2\text{O}$. We have measured Fe-57 and Gd-155 Mossbauer spectra for the new compound. Mossbauer parameters for this compounds are associated with the geometries around Fe(III) and Gd(III) atoms in the coordination polymer structure.

1P28 Development of an apparatus to concentrate ^{188}Re solutions by a multi column system for cancer therapeutic experiments

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Rhenium-188 is a useful generator-produced radioisotope for therapeutic applications because of their energetic beta particles and gamma rays suitable for imaging. Because the ^{188}W parent is obtained in a relatively low specific activity from the irradiation of enriched ^{186}W in a reactor, relatively large volumes of normal saline are required to elute the ^{188}Re from alumina based $^{188}\text{W}/^{188}\text{Re}$ generators. In this study, a simple apparatus for effective concentrations of ^{188}Re solutions from alumina-based $^{188}\text{W}/^{188}\text{Re}$ generators was developed by a tandem cation (IC-H or OnGuard II H) – anion (QMA SepPak) column system. Ammonium acetate was used for the elution of ^{188}Re from the $^{188}\text{W}/^{188}\text{Re}$ generator. Because the ammonium cations are exchanged for hydrogen ions through the first cation column, ^{188}Re was adsorbed to the QMA SepPak without interfering from acetate anion. Rhenium-188 was eluted from the QMA SepPak column with of normal saline after washing with water. More than 95% of ^{188}Re was recovered in 1 ml of saline.

Concentration ratios greater than 50:1 can be achieved with this apparatus.

1P29 Production of RI for Charged Distribution at the RIKEN AVF Cyclotron

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In October 2007, we started a charged distribution of the purified radioisotopes (RIs) of ^{65}Zn and ^{109}Cd to the general public with the collaboration of Japan Radioisotope Association. These RIs were produced by the $^{65}\text{Cu}(p,n)^{65}\text{Zn}$ and $^{109}\text{Ag}(p,n)^{109}\text{Cd}$ reactions with a 14-MeV proton beam from the RIKEN AVF cyclotron. After the irradiation, ^{65}Zn was chemically separated from the target material by the anion exchange in a hydrochloric acid system. Cadmium-109 was separated in the same method as ^{65}Zn after a AgCl precipitation. The impurities as well as the specific activities of the purified RIs were evaluated using ICP-MS. As an example, the specific activities of ^{65}Zn and ^{109}Cd were 410 and 340 kBq/ng, respectively.

In parallel with the ^{65}Zn and ^{109}Cd productions, we are producing short-lived RIs with a gas-jet system. As a test experiment, oxide targets of ^{89}Y , ^{nat}Zr , and ^{141}Pr (about 100 $\mu\text{g}/\text{cm}^2$ each) electrodeposited on Al backing foils (10 μm) were irradiated with the proton beam. The recoiling RIs adsorbed on KCl aerosols were transported with a He carrier gas to a chemistry laboratory, and were collected on a glass filter (ADVANTEC GB-100R). With the 11- μA beam irradiation for 60 s, 110, 130, and 43 kBq of $^{89\text{m}}\text{Zr}$, $^{90\text{m}}\text{Nb}$, and

$^{141\text{m}}\text{Nd}$, respectively, were obtained.

1P30 The biodistribution of Riken Multitracer (MT) and MT-EDTMP, MT-DOTMP in mice

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Samarium-153 labelled ethylenediamine-tetramethylenephosphonic acid (EDTMP) and Holmium-166 labelled

1,4,7,10-tetraazacyclododecane-1,4,7,10-tetramethylenephosphonic acid (DOTMP) have been used for bone pain palliation for bone metastases and myeloablative treatment for multiple myeloma respectively in nuclear medicine. Both phosphonates typically accumulated in bone. The usage of these radiolabelled phosphonates is depending on the half-life, radiation energy, and decay mode of selected nuclides. In order to examine the practical applicability of labelled phosphonates, it is important to investigate their metabolism. The Riken multitracer (MT) technique, which is highly efficient for comparison of many tracers under same conditions, has been used to investigate the biodistribution of MT, MT labelled EDTMP and DOTMP in vivo. Male 8-week-old male ICR mice were grouped and administered with MT (in saline solution), MT-EDTMP, and MT-DOTMP. The blood, liver, kidney, small intestine, large intestine, femur, and brain were excised and weighed. The uptake rates of several nuclides (^{57}Co , ^{65}Zn , ^{75}Se , ^{83}Rb , ^{88}Zr , ^{88}Y , ^{101}Rh) in tissues were determined by gamma-ray spectrometry. The skeletal uptakes of MT-phosphonates except for ^{75}Se and ^{83}Rb were higher than those in other soft tissues. The ^{75}Se and ^{83}Rb showed the same behavior to those in MT. Although the skeletal uptakes of ^{88}Y phosphonates were decreased comparing to that in MT saline solution, the bone to tissue uptake ratios were increased. ^{88}Zr phosphonates were found to show fast uptake and rapid clearance from soft tissues.

2A01 Vertical profiles of ^7Be , ^{137}Cs , ^{226}Ra , ^{228}Ra and ^{228}Th activities in water samples from the Sea of Japan

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To study the vertical transports of water masses and particle matters within the Sea of Japan, a total 81 of water samples were collected from water columns in the Japan Basin (3700 m depth), the Yamato Basin (2610 m), the Tsushima Basin (1161 m), the Yamato Rise (367 m), and the coastal areas along the Japan Islands (65-475 m) in early summer of 2005-2008. By employing a low-background γ -spectrometry combined with simple coprecipitation processing to small volumes (20-60 L) of these seawater samples, ^{137}Cs ($t_{1/2} = 30.2$ y), ^{226}Ra (1600 y), ^{228}Ra (5.75 y), ^{228}Th (1.91 y), and ^7Be (53.3 d) were determined.

All of these nuclides exhibit quite different trends of vertical profiles (e.g., activity levels, activity ratios, depth of maximum activity, and dissolved/particle activity ratio) in each site, suggesting interesting features as for the vertical and lateral movements of water masses, the removal mechanisms of these nuclides and transport of particle matters within the Sea of Japan.

2A02 $^{238}\text{U}/^{230}\text{Th}$ and $^{226}\text{Ra}/^{230}\text{Th}$ activity ratios in rhyolite from Higashi-Izu monogenic volcano group and Izu arc

volcanoes

TAKAHASHI, M., KURIHARA Y., SATO, J.
(Meiji Univ.)

Observation was made with rhyolitic volcanic rocks erupted from Kozu-shima, Niijima and Higashi-Izu monogenic volcano group for activity ratios of $^{238}\text{U}/^{230}\text{Th}$ and $^{226}\text{Ra}/^{230}\text{Th}$. Magma genesis at the subducting zone is generally inferred to be induced by the melting of the mantle wedge by the addition of the fluid derived from the subducting slab. Uranium and radium are highly mobile elements in aqueous fluids relative to thorium, the excess of ^{238}U and ^{226}Ra over ^{230}Th in the arc volcanic rocks has generally been ascribed to the addition of U- and Ra-rich fluids derived from the subducting slab by dehydration. While the activity ratios of basaltic products from Izu-arc showed $^{238}\text{U}/^{230}\text{Th} > 1$ and $^{226}\text{Ra}/^{230}\text{Th} > 1$, the majority of rhyolitic products was in equilibrium.

2A03 Deposition of organic iodine to the soil in Yoro area

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(Grad. School Sci., Hiroshima Univ.,¹ Spring-8/JASRI²)

Environmental behavior of iodine is of great importance related to the release of radioiodine from processing of nuclear fuel, etc. To understand the fate of radioactive iodine in a soil-water system, it is necessary to know the iodine species both in soil and water. Soil and soil water samples were collected at depths of 0, 3, 6, 9, and 12 cm from surface in Yoro area, Chiba, Japan. Iodine species in the soil and soil water were determined by XANES and HPLC-ICP-MS, respectively. Elemental mapping of iodine in soil particle was obtained by micro-XRF at Spring-8 BL37XU.

The depth profile of iodine concentration in soil correlates quite well with that of organic carbon content. XANES spectra of soil samples were well fitted by the spectrum of SRHA (Suwannee River humic acid). Elemental mapping shows that iodine is incorporated in organic matter rather than in iron oxides. Accordingly, most of iodine in soil is combined with organic matter such as humic substances. In soil water, 40% of organic iodine and 60% of I^- were detected at 3-6 cm depth, while organic iodine is only several percent at 9-12 cm. Organic iodine plays an important role in the fixation of iodine in soil.

2A04 Effect of complexation with humic substances and EDTA on the diffusion of metal ions in water

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(Grad. School Sci., Hiroshima Univ.)

In this study, the diffusion coefficients of free metal ions (M) and their complexes with HS (M-HS) were compared to understand the effect of complexation with HS on the diffusion of metal ions such as Co, Cd, and rare earth elements (REE). Although the diffusion coefficients of free metal ions depend on ionic potential, such dependence was not observed in the presence of HS. Comparing the diffusion coefficients of metal complexes with ethylenediaminetetraacetate (EDTA), fulvic acid, and humic acid showed that the molecular weight (MW), or the size of the ligand, is of primary importance for the diffusion of M-HS. As a consequence, the diffusion coefficients of every REE were similar in the presence of HS, while they were different in the absence of HS due to the different size of each REE. The similarity among the diffusion coefficients of REE-HS was caused by the much larger size of HS compared with each ion. However, the distribution coefficients of M-HS were not similar among REE,

Cd, and Co. REE and Cd which have higher affinities for larger MW fraction in HS diffused slower than Co which favors smaller MW fraction. The results show that the affinity for different MW fractions among HS controls the diffusion of M-HS.

2A05 Studies on the analysis of ^{129}I by AMS and its application to iodine rich brine samples

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(Faculty of Science, Gakushuin Univ.,¹ MALT, University of Tokyo²)

Brine samples with high iodine concentrations are found in several places in Japan. It is interesting to note that iodine is often associated with methane. However, details on the iodine accumulation process and origin of the brines are not well known. Therefore, we have tried to use ^{129}I as a tracer to discuss geochemical behavior of iodine. For the analysis of $^{129}\text{I}/^{127}\text{I}$ ratios, iodine fraction was extracted into CCl_4 as I_2 by the addition of NaNO_2 . It was back-extracted into aqueous solution containing Na_2SO_3 as iodide. Finally, iodide was precipitated as AgI . Then the ratio of $^{129}\text{I}/^{127}\text{I}$ was determined by AMS at MALT facility of the University of Tokyo. $^{129}\text{I}/^{127}\text{I}$ ratios in brine samples collected from Chiba prefecture were successively determined. The values obtained from 7 determinations at MALT were $(0.173 \pm 0.022) \times 10^{-12}$, which agreed well with our previous data measured at Purdue University. We are going to introduce our analytical results on stable and ^{129}I in variety of brine samples.

2A06 Isotopic shifts caused by interaction of cosmic rays with planetary surface materials

HIDAKA, H.
(Grad. School Sci., Hiroshima Univ.)

Thermalized neutrons arise from the interaction of high-energy cosmic rays (galactic cosmic rays: GCR) with planetary surface materials. One of the most significant reactions with neutron is neutron capture reaction of the (n, γ) type. Because ^{149}Sm , ^{155}Gd and ^{157}Gd have extraordinarily large thermal neutron capture cross sections, isotopic shifts of $^{150}\text{Sm}/^{149}\text{Sm}$, $^{156}\text{Gd}/^{155}\text{Gd}$ and $^{158}\text{Gd}/^{157}\text{Gd}$ resulted from the neutron capture reactions $^{149}\text{Sm}(n, \gamma)^{150}\text{Sm}$, $^{155}\text{Gd}(n, \gamma)^{156}\text{Gd}$ and $^{157}\text{Gd}(n, \gamma)^{158}\text{Gd}$, respectively, can be sometimes detected in meteorites and lunar materials. The production rate of neutron depends upon the depth of the sample in a large object (or size of an object) and chemical composition of the target material. Therefore, Sm and Gd isotopic shifts are used to characterize the cosmic-ray exposure records of planetary materials. In this talk, I would like to present two topics on the neutron capture reactions of lunar regoliths and meteorites studied from $^{150}\text{Sm}/^{149}\text{Sm}$ and $^{158}\text{Gd}/^{157}\text{Gd}$ isotopic shifts.

2B01 Magnetic property of the spin-crossover iron(III) compound with structural phase transition

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Generally, spin-crossover (SCO) iron(III) compounds exhibit gradual SCO behaviors compared with SCO iron(II) compounds. However, some SCO iron(III) compounds exhibit abrupt SCO behaviors with thermal hysteresis loop. The abrupt

SCO behaviors with hysteresis loop are corresponding to a first order phase transition. For such the compounds, cooperativities arising from the strong intermolecular interactions exist in the compounds. Our objective is to design strongly cooperative SCO assemblies consisting of mononuclear molecules with strong intermolecular interactions as such π - π stacking or hydrogen bonding, and to find out the photoswitchable molecules in the compounds with bistability. Here we report about LIESST effect for spin transition iron(III) compounds [Fe(acpa)₂]PF₆ (**1**), [Fe(acpa)₂]BF₄ (**2**) and [Fe(acpa)₂]ClO₄ (**3**) (acpa = *N*-(1-acetyl-2-propylidene)-2-pyridylmethylamine). The compounds **1** and **2** exhibit gradual SCO behavior at $T_{1/2}$ = 200 K and 290 K without hysteresis loop. On the other hand, the compound **3** exhibits unique magnetic behavior. Before annealing at 250 K, the compound **3** exhibits gradual SCO behavior in the temperature range of 260 K – 400 K. However, after annealing at 250 K, the compound **3** exhibits abrupt spin transition at $T_{1/2}\uparrow$ = 380 K and $T_{1/2}\downarrow$ = 365 K with hysteresis loop.

2B02 Structural change and spin-crossover switching triggered by adsorption and desorption of organic molecules for the assembled iron complexes bridged by 1,3-bis(4-pyridyl)propane

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We synthesized assembled complexes Fe(NCX)₂(bpp)₂ (X = S (**1**), Se (**2**), and BH₃ (**3**)) (bpp = 1,3-bis(4-pyridyl)propane). **3** has a novel 2D interpenetrated structure and shows spin-crossover phenomenon. We synthesized Fe(NCBH₃)₂(bpp)₂·2(benzene) (**4**) which is a benzene-enclathrated complex of **3**, and studied the magnetic behaviors and structural properties by using Mössbauer spectroscopy and so on.

4 is in a temperature independent high-spin state. By releasing benzene in the air, **4** becomes Fe(NCBH₃)₂(bpp)₂. This guest-free **4** is low-spin state at low temperatures, and indicates a spin-crossover phenomenon. The guest-free **4** enclathrates benzene molecules reversibly by setting in the benzene atmosphere, returned to the temperature independent high-spin state. Therefore, **4** displays spin-crossover switching triggered by adsorption and desorption of organic molecules. We succeeded to follow a series of spin states change by Mössbauer spectroscopic measurements.

We revealed that the spin-crossover switching in **4** is accompanied by the change of assembled structure. Zn(NCBH₃)₂(bpp)₂·2(benzene) (**4'**) with the same structure as **4** has 1D chains structure. This result and powder X-ray diffraction patterns indicated the change of assembled structure; Fe(NCBH₃)₂(bpp)₂·2(benzene) [1D chains] \leftrightarrow Fe(NCBH₃)₂(bpp)₂ [2D interpenetrated structure]. Thermal study of this change and magnetic behaviors of clathrate complex including guests except for the benzene are ongoing.

2B03 ¹⁹⁷Au Mössbauer spectra of cyclometalated dinuclear gold compounds having 2-C₆F₄PPh₂

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(Dep. of Sci., Toho Univ.¹ School of Applied Sciences, RMIT Univ.,² Research School of Chem., Australian National Univ.³)

Gold-197 Mössbauer spectra of cyclometalated digold(I) compound [Au₂(μ -2-C₆F₄PPh₂)₂] (**1**) and its derivatives

[Au₂X(μ -2-C₆F₄PPh₂)₂] (**2**) and [XAu(μ -2-C₆F₄PPh₂)(κ^2 -2-C₆F₄PPh₂)AuX] (**3**) are measured at 20 K using a ¹⁹⁷Pt/Pt source. The spectra for **1** and **2** indicate the presence of equivalent gold atoms, whereas spectra for **3** show absorptions due to two inequivalent gold centres. The plot of quadrupole splitting against isomer shift indicate that the gold-phosphorus bond in these compounds is less covalent than the gold-phosphorus bond in protio and methyl analogues.

2B04 Development of in-beam Mössbauer spectroscopy using an RI beam in HIMAC of NIRS

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The possibility of the on-line Mössbauer spectroscopy combined with ion implantation using a radioactive ⁵⁷Mn ($T_{1/2}$ = 1.45 min) beam in HIMAC of NIRS has been considered. HIMAC is one of the few accelerators that are able to supply an energetic RI beam such like ⁵⁷Mn. The ⁵⁷Mn was produced by a projectile-fragmentation reaction of a ⁵⁹Co (E = 500 MeV/nucleon) primary beam impinging on a 25 mm thick Be production target. The typical intensity of ⁵⁷Mn produced in HIMAC was estimated to be 9.5×10^4 particle/s, being one digit less than the amounts obtained by the RIPS in RIKEN Accelerator Research Facility. The ⁵⁷Mn nuclei were implanted into a 1 mm thick Al plate, and were stopped using a Pb energy-attenuator with an appropriate thickness. The γ -ray spectra were measured by a Ge detector, and a ⁵⁷Mn implantation Mössbauer spectrum was accumulated by a gas-filled resonance counter (PPAC), respectively. These obtained results suggested that it would be possible to perform the in-beam Mössbauer spectroscopy using a ⁵⁷Mn beam in HIMAC.

2B05 Local fields in In-doped ZnO observed by means of the TDPAC method

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Zinc oxide is an intrinsic *n*-type semiconductor, and it is known that its conductivity varies with the presence of dilute dopants. In order to compare and/or contrast physical properties observed for the bulk sample with microscopic information, we applied the time-differential perturbed angular correlation method with the ¹¹¹Cd(\leftarrow ¹¹¹In) probe to a study of local fields in 0.05 at.% Indium-doped and undoped ZnO. For the In-doped ZnO sample, temperature dependence was observed for the amplitude and electric quadrupole frequency of the main component in the TDPAC spectra; whereas this phenomenon was not observed for the second component. In addition, the ratio of the two components changes with temperature. In the present talk, we discuss the probe sites and the density of conduction electrons at the probe on the basis of the temperature dependence of the TDPAC spectra.

2B06 Measurement of hyperfine field in metal complexes and mavycyanin by PAC method using ¹¹¹Ag probe (II)

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School Sci., Osaka Univ.⁴, Res. Reactor Inst., Kyoto Univ.⁵.)

A parent PAC probe of ¹¹¹Ag, which has a half life of 7.45d was introduced to TDPAC measurements in order to seek a better condition for biomolecular study. In the previous work, we succeeded in measuring the TDPAC spectra of various complexes and wild-type mavycyanin by using the ¹¹¹Ag probe, and obtained their electric field gradients. In this study, the complex samples with ligands of quinaldate, DDC, oxine and BHPA were prepared and subject to PAC measurements. The electric field gradient values obtained from the TDPAC spectra were compared with the data obtained with other probes of ¹¹¹In, ^{111m}Cd and ¹¹⁷Cd. The probe dependence of electric field gradient was discussed from the result.

3A01 Studies on hot atom chemical behavior of energetic ions in solids (XVI) -Retention behavior of hydrogen isotopes in the tungsten - carbon mixed layer-

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In D-T (deuterium-tritium) fusion reactors, the combined usage of tungsten (W) and carbon (C) has been considered as the divertor materials to endure the high heat flux from the plasma. Under this condition, some of C will be ionized and implanted into W with energetic hydrogen isotopes including T. Therefore, it is expected that the W - C mixed layer would be formed on the surface of divertor during discharged and the chemical behavior of energetic T implanted into the W - C mixed layer will be required.

In this study, C⁺ and D₂⁺ were simultaneously and sequentially implanted into W and effects of C implantation on the retention behavior of D were investigated by means of TDS and XPS. From the TDS spectra, broad desorption peak was found at 800 K for sequentially implanted W. It was suggested that the desorption peak around 800 K was contributed to C-D bond because it was reported in our previous study [1]. On the other hand, no C-D bond was observed for simultaneous implantation due to higher sputtering rate by ions.

[1] H. Kimura, *et al.*, *Fusion Eng.Des.* **2006**, *81*, 295.

3A02 Studies on hot atom chemical behavior of energetic ions in solids (XVII) - Dependence of implantation temperature on chemical behavior of energetic hydrogen isotopes implanted into carbon-contained boron films -

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(Radiosci. Res. Lab., Fac. of Sci., Shizuoka Univ.)
For the first wall of D (deuterium)-T (tritium) fusion reactor, boronization has been thought to be applied to remove impurities such as oxygen and carbon. In the fusion environment, energetic particles including tritium would be implanted into the boron film. Therefore, it is important to understand tritium chemical behavior in carbon-contained boron film from the view point of tritium safety. In this study, our attention was paid to the role of carbon on tritium trapping process.

A carbon-contaminated boron film with the carbon concentration of 35 % was prepared by the plasma CVD apparatus, and deuterium ions (D₂⁺) were implanted into the film with various implantation temperatures. The chemical states of carbon and boron were evaluated by XPS and the deuterium desorption behavior was also studied by TDS.

From the D₂ TDS spectrum for the sample implanted at room

temperature, it is found that there were three D₂ desorption stages located around 520 K, 700 K and 950 K. It was reported that these stages were desorption processes of deuterium trapped as B-D-B, B-D and B-C-D bonds, respectively. Deuterium retentions as B-D-B and B-D bonds were decreased with increasing of implantation temperature, while that as B-C-D bond was constant up to 673K. These results indicate that the formation of B-C-D bond would be induced by a non-thermal equilibrium process.

3A03 Studies on hot atom chemical behavior of energetic ions in solids (XVIII) - Annihilation behavior of irradiation defects induced in 14 MeV neutron-irradiated lithium ortho-silicate-

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(Radiosci. Res. Lab., Fac. of Sci., Shizuoka Univ.¹, Japan Atomic Energy Agency²)

In blanket systems for D-T fusion reactors, lithium compounds play a role of tritium breeding. Lithium ortho-silicate (Li₄SiO₄) is one of candidates for tritium breeding materials due to its high lithium density and low radioactivation. From the viewpoint of the establishment of tritium recovery techniques, it is important to elucidate the chemical behavior of tritium produced in Li₄SiO₄. In this study, the annihilation behavior of irradiation defects in 14 MeV neutron-irradiated Li₄SiO₄ was investigated by means of Electron Spin Resonance (ESR). These results were compared with those of thermal neutron-irradiated sample.

From the results of isothermal annealing experiments, it was assumed that the annihilation process consisted of two processes, namely, fast and slow processes. From the Arrhenius plot of annihilation rate of irradiation defects, the activation energy for each process in 14 MeV neutron irradiated sample was determined. From the comparison with slow processes for thermal and for 14 MeV irradiated samples, the activation energy for thermal neutron-irradiated sample was larger than that for 14 MeV neutron-irradiated sample, indicating that, the density of irradiation defects influence on oxygen trapped at interstitial site.

3A04 Long-term variability of plutonium and thorium isotope depositions observed in Tsukuba

HIROSE, K., IGARASHI, Y., AOYAMA, M.
(Geochem. Res. Dep, Meteor. Res. Inst.)

Monthly depositions of plutonium and thorium isotopes in Tsukuba, Japan during the period 1990-2007 were determined. The monthly ^{239,240}Pu deposition showed a typical seasonal variation with inter-annual variability. Especially, marked peaks of the ^{239,240}Pu deposition occurred in spring in the 2000s. The seasonal change and inter-annual variation of the monthly ^{239,240}Pu deposition are closely related to those of the Kosa events. High monthly depositions of thorium, which originates from soil particles, were observed in spring as did ^{239,240}Pu. The ²³⁰Th/²³²Th activity ratios in deposition samples showed large variability; its higher ratios were observed in early spring. The higher ²³⁰Th/²³²Th ratios reflect local soils contaminated by uranium originating from fertilizers, whereas the low ratios occur in lithogenic materials. Therefore, the ²³⁰Th/²³²Th ratios in deposition allow us to separate local and remote fractions of the thorium deposition. The results reveal that both local and remote fractions of the thorium deposition showed seasonal variations with high in spring although the remote fraction is

dominant within season. We calculated the local fraction of the $^{239,240}\text{Pu}$ deposition when constant $^{239,240}\text{Pu}/^{232}\text{Th}$ ratio is presumed in local soil particles. The local $^{239,240}\text{Pu}$ deposition showed a smaller peak in early spring, corresponding to local dust storm event. The marked high $^{239,240}\text{Pu}$ deposition in spring in the 2000s is attributable to remote fraction, corresponding to the Kosa.

3A05 Distribution of ^{129}I in the surface soil at Rokkasho, Aomori, Japan

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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 have determined ^{129}I concentrations in uncultivated soil and various cultivated soils at Rokkasho by AMS, for evaluation of the effects of the release on the environmental ^{129}I concentration level. Concentration of ^{129}I in the uncultivated soil surface showed *ca.* 1.4 mBq kg⁻¹-dry soil, and $^{129}\text{I}/^{127}\text{I}$ isotopic ratio showed 10⁻⁸.

This work was performed under contract with the Aomori Prefectural Government, Japan.

3A06 Radiocarbon dating of the *kohitsugire* calligraphies and the *kiwamefuda* certificates: On the execution age of *Genji* narrative scroll

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(Nagoya Univ.,¹ Chuo Univ.²)

Many ancient manuscripts written in *Heian-Kamakura* period (9th - early 14th century) had been divided into leaves to appreciate as hanging scrolls. Such ancient paper sheets with elegant calligraphy are called *kohitsugire*. In this study, radiocarbon ages of *kohitsugire* calligraphies attributed to *Asukai Masatsune* (1170-1221), *Imaki-gire* and *Kinginkirihaku-wakanroaisyu-gire*, were measured by AMS. Although they are attributed to *Asukai Masatsune* according to the *kiwamefuda* certificates, it has been commonly accepted in recent years that they are genuine handwritings of *Fujiwara no Norinaga* (1109-1180?). Radiocarbon dating indicated that they were written from the middle 11th to the late 12th century. It includes the period when *Fujiwara no Norinaga* had flourished as a calligraphist and is too early as *Asukai Masatsune's* handwriting. The national treasure "*Genjimonogatari emaki*" is a narrative scroll depicting *Murasaki Shikibu's* novel "Tale of Genji" by colorful paintings and graceful calligraphy. A part of the calligraphy in "*Genjimonogatari emaki*" is written by the same handwriting as *Imaki-gire* and *Kinginkirihaku-wakanroaisyu-gire*. The radiocarbon ages of these *kohitsugire* shows that they were written by *Fujiwara no Norinaga*. Therefore, the result also suggests that "*Genjimonogatari emaki*" was executed in the late 12th century.

3A07 Oxidation of nobelium by an electrochemical approach

TOYOSHIMA, A.,¹ KASAMATSU, Y.,¹ TSUKADA, K.,¹ KITATSUJI, Y.,¹ ISHII, Y.,¹ TOUME, H.,¹ ASAI, M.,¹ NAGAME, Y.,¹ HABA, H.,² AKIYAMA, K.,³ OOE, K.,⁴ SATO, W.,⁴ SHINOHARA, A.⁴

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We will present oxidation of nobelium (No) using an electrochemistry apparatus combined with a chromatographic separation technique available for single atoms. Nobelium-255 (3.1 min) was produced in the $^{248}\text{Cm}(^{12}\text{C},5n)$ reaction at the JAEA tandem accelerator. Chromatographic behavior of No on a chemically modified electrode with Nafion perfluorinated ion-exchange resin was investigated in 0.1 M α -hydroxyisobutyric acid (α -HIB) solution. Elution behavior of ^{81}Sr and ^{162}Yb produced by the $\text{Ge}(^{12}\text{C},xn)$ and $\text{Gd}(^{12}\text{C},xn)$ reactions, respectively, was also measured in separate experiments under the same conditions as those of ^{255}No to verify the difference in elution behavior between the divalent (Sr^{2+}) and trivalent (Yb^{3+}) ions. Independently of the applied potentials, $^{162}\text{Yb}^{3+}$ was eluted in 0.1 M α -HIB while $^{81}\text{Sr}^{2+}$ strongly adsorbed on the electrode. At the low applied potential of 0.2 V, ^{255}No was adsorbed on the electrode, indicating that No is bound in the most stable divalent state. On the other hand, at the higher potential of 1.2 V, ^{255}No was eluted with 0.1 M α -HIB, showing that No exists as a trivalent ion. These results demonstrate that No^{2+} is successfully oxidized to No^{3+} with the present electrochemical approach.

3A08 Cross section measurements for monoenergetic neutron-induced nuclear reaction of Cu and Nb in the intermediate energy region.

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(Grad. School Sci., Osaka Univ.,¹ Res. Reactor Inst., Kyoto Univ.,² KEK,³ RCNP, Osaka Univ.,⁴ Space Sci. Lab., California Univ.⁵)

Neutron induced nuclear reaction has no coulomb barrier. Low energy nuclear reactions induced by the neutrons are different from those by the protons because of large influence of the coulomb barrier. On the other hand, high energy nuclear reactions by the neutrons are similar to those by protons due to negligible effect on the coulomb barrier. However, there are no experimental data of the reaction cross sections above 100 MeV neutrons. In this study, we tried to determine the cross sections of the reaction products in copper and niobium for intermediate energy (several hundred MeV) monoenergetic neutrons. 362 MeV neutrons were produced by bombarding the ^7Li target with the 392 MeV proton beam at the N0 course in Research Center for Nuclear Physics (RCNP), Osaka University. The copper and niobium foils were also irradiated directly with the proton beam to compare the results of neutron induced nuclear reactions. The produced radionuclides were detected with a gamma-ray counting method by using high purity germanium detectors. We obtained the cross sections of 24 nuclides for neutron-induced nuclear reactions in copper, and those of 31 nuclides in niobium. In this presentation, we report the obtained neutron induced cross sections of copper and niobium, and compare the results with the previous ones in literature to discuss the differences between neutron and proton induced nuclear reactions.

3A09 Liqui-liquid extraction behaviors of Nb and Ta for Db chemistry

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In order to investigate the chemical property of ¹⁰⁵Db, the liquid-liquid extraction behavior of Nb and Ta with the quaternary ammonium salt Aliquat 336 in pure HF and HCl systems has been studied. The liquid-liquid extraction experiments were performed with a batch method. The Aliquat 336-1,2-dichloroethane solution and the various HCl or HF solution containing the radiotracer were shaken in the Teflon vessel. After centrifuging, an activity of each phase was measured and a distribution coefficient was calculated from the ratio of the activity of the organic phase to that of the aqueous phase. The distribution coefficients of Ta in the Aliquat 336/HF system were very high as well as those in the previous work by other group.

On the other hand, the extraction of Ta from HCl was very weak, which was different from the previous results. The reason for this difference in HCl system may be adsorption of a part of Ta tracer to the polypropylene vessel used in the previous experiments.

[1] W. Paulus, J. V. Kratz, E. Strub, and S. Zauner, *Radiochim. Acta* **84**, 69-77 (1999)

3A10 Solvent Extraction of Mendelevium with HTTA in Carbon Tetrachloride from Hydrochloric Acid

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(Grad. School Sci., Osaka Univ.¹, RIKEN², JAEA³)

The chemical characters of f-block elements have been studied by comparing the distribution ratio (D) of liquid-liquid extraction for the rare earth elements and the actinide elements systematically. In this study, the extraction behavior of mendelevium (Md) into 0.1 M 2-thenoyltrifluoroacetone (HTTA)-CCl₄ from 0.1 M NH₄Cl-HCl has been studied together with that of thulium (Tm).

²⁵⁵Md (T_{1/2} = 27 min, E_α = 7.327 MeV) and ¹⁶²Tm (T_{1/2} = 21.7 min, E_γ = 900 keV) were produced by irradiating a mixed target (²⁴⁸Cm 932 μg/cm², ^{nat}Gd 84.3 μg/cm²) with a 56.3 MeV ¹¹B beam at K70 AVF cyclotron in RIKEN. Reaction products were carried to the chemistry laboratory by the gas-jet system, collected on a polyester film, and dissolved in 0.1 M NH₄Cl-HCl with either pH 3.05, 3.16, or 3.36. The solution and the same volume of 0.1 M HTTA-CCl₄ were placed in a polyethylene tube and shaken for 20 minutes. The each phase was isolated, dried up on tantalum dishes, and subjected to α-particle spectrometry.

As a result, the slopes of log D vs. pH plot were 1.42±0.18 for Md and 1.74±0.05 for Tm. The slopes provide the number of separated protons from HTTA when HTTAs were coordinated to the metal ion. However three HTTAs were coordinated to actinide and lanthanide in literature. In our study, we can not rule out the possibility that the large amount of NH₄Cl affected to the experimental results.

3A11 Observation of etch pit shape in CR-39 with atomic force microscope for the samples irradiated with 25 MeV C-12 ions

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

In this study, we aim to acquire a standard data of the relation between an incident particle and its etch pit shape for particle identification with a solid-state track detector. The irradiation experiment of ¹²C ion was done at the Wakasa wan energy research center. The irradiated sample was etched for 15 minutes with 6M NaOH solution, and the etch pit was observed with an atomic force microscope. The depth of etch pit and the dimension of its opening were examined in relation to the incident energy of ¹²C ion. The depth of etch pit becomes shallower as the incident energy increases. And, the opening becomes smaller as the energy increases. The behavior agrees with the expected stopping power depending on the incident energy of ions. However, it is considered necessary to examine etching condition etc. because there is a considerable deviation of the shape even in a sample under the same conditions.

3B01 Evaluation of neutron activation in medical accelerator facilities

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(High Energy Accelerator Research Organization)

The definition and handling of activated material at the accelerator facilities have been just proceeded to adopt in Japanese regulations. Neutron is a major source of activation in surrounding materials of small accelerators, such as electron linear accelerators and cyclotrons. In order to evaluate the activation levels by monitoring neutron flux during operation, we used personal dosimeter (TLD, CR-39) and activation foils. It was found that the activation method using Au foils is sensitive for thermal neutrons. Activity observed in bolts obtained in the accelerator room is suited for analyzing the historical record of activation. CI-36 AMS is one of the most sensitive method for the measurement of thermal neutron fluence in concrete samples. On the other hand, several Monte Carlo codes are also applied to estimate neutron propagation and neutron activation. It was confirmed these results were consistent with the results of activity measurement.

3B02 Neutron Flux Estimation in a Cyclotron Room by Gamma-ray Analysis of Bolts

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(Nagoya Univ.¹, Nagoya Univ. Grad.², Longevity Sci.³, KEK⁴, Nagoya Rehabilitation c.⁵)

Short half-life nuclides for PET examination are mainly produced using on site small cyclotrons. Operation of the cyclotron inevitably generates significant quantity of neutrons, which will potentially activate the cyclotron and surrounding materials. We tried to estimate the neutron fluxes in cyclotron rooms via the radioactivity analysis of bolts on wall sockets in the rooms. The objectives in this study were two cyclotrons; one was a negative ion type and the other was a positive ion type, while both cyclotrons were able to accelerate protons and deuterons up to 18 MeV and 10 MeV, respectively. The bolts on the wall sockets in the cyclotron rooms were removed and the radioactivities were analysed by an HPGe detector. The results were compared with those measured via the gold foil activation method. Main nuclides detected in the bolts were ⁶⁴Cu and ⁶⁵Zn. The neutron fluxes of both cyclotron rooms

were from $4 \cdot 10^5$ to $2 \cdot 10^6$ $\text{cm}^{-2}\text{s}^{-1}$. The neutron fluxes estimated from the activities of ^{64}Cu and ^{65}Zn in the bolts approximately agreed with ones measured via the gold foil method. It was proved that the radioactive analysis of those bolts is effective to estimate the neutron flux in cyclotron rooms.

3B03 Determination of neutron fluxes at the self-shielded PET cyclotron and the electron linac apparatus of Tokushima University Hospital using activation foil method

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At present, from a radiation safety management point of view, it has been discussed to construct guidelines on estimation of neutron fluxes produced via operating various medical small accelerators and to establish uniformly the clearance system related to having neutron activation effects into each accelerator facility. That is, it was the aim of this investigation to accumulate the data on estimation basis of neutron flux measurements at those medical small accelerators by an activation foil method. In this work, the neutron fluxes at the self-shielded PET cyclotron and the electron linac apparatus of Tokushima University Hospital have been measured as the medical small accelerator. As a result, for the self-shielded PET cyclotron, it was found that the thermal neutron flux is $(1.04 \pm 0.05) \times 10^7$ $\text{cm}^{-2} \text{s}^{-1}$ and the fast neutron fluxes are distributed over the range 1×10^4 (the neutron energy $E_n = 9$ MeV) to 1×10^7 ($E_n = 1.5$ MeV) $\text{cm}^{-2} \text{s}^{-1}$ into the radiation shelter, and while the thermal neutron fluxes would be distributed over the range 5.0×10^1 to 9.9×10^1 $\text{cm}^{-2} \text{s}^{-1}$ and then the fast neutron dose leakages were not detected outside there. For the 6/10MV electron linac apparatus, it was also found that the same thermal neutron fluxes would be distributed all around the linac room and the measured values are over the range $(1.01 \pm 0.18) \times 10^3$ to $(1.32 \pm 0.10) \times 10^3$ $\text{cm}^{-2} \text{s}^{-1}$.

3B04 Process of off-gas generated from the spallation neutron source (JSNS) in J-PARC

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Various kinds of the spallation products are generated in the mercury used as the spallation neutron source (JSNS) at the Materials and Life Science Facility in J-PARC. In particular, gas products are accumulated in the gas phase of the mercury tank in the mercury circulation system. An off-gas processing system has been installed in order to process the off-gas and enable the gas release to the environment under the concentration limit of radioactivity. The off-gas processing system has three functions mainly as follows: (1) Removal of mercury vapor by a mercury absorption filter, (2) Removal of tritium by a tritium process circulation system, and (3) Reduction of noble gas activities by keeping the gas using a gas holder for about one year. In this presentation, we report the calculation of the amount of the radioactive gas product in the mercury, which is used for the system design as a basis, outline

of the off-gas process and the activity measurement of the off-gas after the commissioning of JSNS with low power operation.

3B05 A novel production method of no-carrier-added ^{64}Cu using chelate exchange method for medical applications

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^{64}Cu has intermediated half-life and multiple decay mode ($\beta^+ = 19\%$, $\beta^- = 39\%$, EC = 42%). It has been expected to apply to medical applications. We have produced no-carrier-added ^{64}Cu via the $^{64}\text{Ni}(p,n)^{64}\text{Cu}$ reaction, and we developed automated apparatus for its production. Generally, an anion exchange method has been employed in the production. However, it is difficult to separate radio-cobalt which is produced by irradiation of enriched-Ni target. The anion exchange method requests a long column and much solvent in order to separate ^{64}Cu with high radionuclide purity. Then, it is necessary to improve the separation method for automated apparatus. In this study, we develop a novel production method using chelate exchange separation for medical applications. As a result, we could separated no-carrier-added ^{64}Cu with high yield (88%) and radionuclide purity (>99%). Obtained ^{64}Cu by this method has low impurities and high specific activity (655 GBq/ μmol), and ^{64}Cu labeled antibodies can be synthesized with high labeling yield (86%). These results indicate that the novel production method can be applied to automated apparatus for medical applications.

3B06 Emission of CH_3T , HT and HTO from Stainless Steel Surface - Effect of Metal Friction and Oxygen -

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We have reported that the emission of methane occurs by the metal-metal collision or friction of stainless steel (SS) by using tritium (T) as a tracer. Since the emission of trace amount water was not detected by the radiogaschromatography (radio-GC), we introduced a chemical HTO trapping column prior to the cryogenic preconcentration column for radio-GC. T_2 gas for tritiation was first introduced into the SS canister which contains many small SS balls inside. The canister was heated and evacuated. The canister was then shaken under the He atmosphere with O_2 . The O_2 content in He and time of shaking were varied as the experimental condition. Emitted CH_3T and HT were trapped cryogenically by the column packed with molecular sieves 13X at liquid nitrogen temperature, and then measured by radio-GC system equipped with a proportional counter. Emitted HTO was trapped by a glass column (1/4-inch o.d., 8-cm long) packed with Sicapent (desiccant agent which consists of mainly P_2O_5), which was then dissolved with water and measured with a liquid scintillation counting system. The trapping efficiency of this column in this experimental condition was confirmed to be nearly 100%. HTO was observed under all the experimental conditions, although CH_3T was emitted only with shaking in absence of O_2 ; HT was emitted only in absence of O_2 .

3B07 Iron carbide films produced by pulsed laser deposition

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YAMADA, Y.,¹
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Laser deposition of Fe in acetylene atmosphere produced iron carbide films having various compositions. The chemical properties of iron carbide films were investigated by Mössbauer spectroscopy and the surface images were observed by scanning electron microscopy. The composition of iron carbide films changed depending on the pressure of acetylene atmosphere (2.7 - 130 Pa) and the temperature of Al substrates (300 K and 573 K). The major component of the films produced at 300 K was found to be a paramagnetic species independent of the pressure of acetylene atmosphere. The paramagnetic iron carbide showed a doublet at both 300 K (I.S. = 0.33 ± 0.02 mm/s, Q.S. = 0.94 ± 0.03 mm/s) and 15 K (I.S. = 0.44 mm/s, Q.S. = 0.95 mm/s) confirming that this species was not magnetic iron oxides, though the Mössbauer parameters do not accord with those of iron carbides reported in literatures. When the films were produced at 573 K, the compositions of the films were different: Fe₃C was produced at 2.7 Pa, and paramagnetic species were produced at high pressure. It was found that iron carbide films having various compositions are produced by changing the substrate temperature and the pressure of acetylene atmosphere.

3B08 Mechanism of formation of Iron oxide thin films produced by laser deposition in oxygen atmosphere

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(Tokyo Univ. of Science,¹RIKEN.²)

Iron oxide films were produced by laser-deposition of Fe in an oxygen atmosphere. The amount of droplets (micrometer-sized Fe particles in Fe vapor) was controlled by shifting the focal point of a Fe metal sample. The compositions and chemical properties of the iron oxide films were studied by Mössbauer spectroscopy and the surface morphology were observed by SEM. An iron oxide film obtained in 7 Pa oxygen atmosphere without droplet was constituted of Fe₃O₄ (two sets of sextet), divalent iron oxide particles (doublet), and trivalent iron particles (doublet). Iron oxide particles were produced by the reaction of laser vaporized Fe atoms with oxygen molecules in gas phase, and the following surface reaction and the crystal growth produced Fe₃O₄ films. On the other hand, the film obtained in 7 Pa of oxygen atmosphere with a large number of droplets in Fe vapor was constituted of Fe₂O₃ (sextet), divalent iron oxide particles (doublet), and *a*-Fe (sextet). In the gas phase reaction, only the surface of Fe droplets was oxidized while the inside of the droplet was not oxidized. Therefore, the particles produced the films consist of a mixture of iron oxides and neat *a*-Fe by deposition onto an Al substrate.

3B09 Synthesis and Mössbauer studies of iron-lanthanoid polynuclear complexes

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We studied the lattice dynamics of the Fe atom in the lanthanide (Ln) complexes with the ferrocenedicarboxylate (L) ligands by ⁵⁷Fe Mössbauer spectrometry. Ln₂L₃ and [Ln(phen)₂]₂L₃ complexes were synthesized and confirmed by the elemental analysis and IR spectrometry. As the result of ⁵⁷Fe Mössbauer measurements, no significant change in the isomer shift (I.S.) and the quadrupole splitting (Q.S.) for both Eu₂L₃ and [Eu(phen)₂]₂L₃ complexes were observed in the temperature range between 80K and room temperature. The

lnA/dT of Eu₂L₃ and [Eu(phen)₂]₂L₃ evaluated from the least square fit of the area intensity for each temperature were -0.0044 and -0.0046, respectively. These values are almost same as each other in spite of the existence of more bulky ligand and are larger than that of the previously reported for the ferrocenedicarboxylic acid. It is found that the molecular motion of L in these complexes are smaller than that of H₂L.

3B10 Mössbauer study of new electrically conductive glass

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*NTA glass*TM, such as 20BaO · 10Fe₂O₃ · 70V₂O₅, is commercially used as a probe for "ionizer (air cleaner)". Electrical resistivity (ρ) of *NTA glass*TM is decreased from the order of $10^6 \sim 10^7 \Omega\text{cm}$ to several Ωcm after heat treatment at a temperature close to the crystallization temperature (T_c). An activation energy for the electrical conduction (E_a) decreases from 0.35 to 0.10 eV after heat treatment. ⁵⁷Fe Mössbauer spectra of *NTA glass*TM show a marked decrease in the quadrupole splitting (Δ) of Fe^{III}O₄ tetrahedra after heat treatment. These results indicate that a structural relaxation of glass network, accompanied by an increase in the local symmetry of distorted FeO₄ and VO₄ tetrahedra, accompanies a decrease in the E_a and that in the band gap. As a result, the number of electrons (polarons) populating the conduction band will be increased.

Heat treatment of LiFeVPO₇ glass also shows a structural relaxation of the network and a marked decrease in the ρ . Capacity of lithium-ion battery could be increased from 50 to 150 mAh g⁻¹ when heat-treated LiFeVPO₇ glass was used as a cathode active material.

3B11 ⁵⁷Fe and ¹¹⁹Sn CEMS of SnO₂ films implanted with ⁵⁷Fe

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The origin of magnetic interactions in Diluted Magnetic Semiconductors (DMS) is attracting a great attention as a basic problem on magnetism[1-2]. In all of these works, the observed ferromagnetism has been attributed to interactions between the magnetic impurities. We have reported the different types of magnetic source in case of Fe doped SnO₂ powder [3], and the main source has never been associated to magnetically ordered defects. The phonon density of states of rutile type structures was studied by NIS [4]. Here we have made the thin films of SnO₂ implanted with ⁵⁷Fe and characterized them by ⁵⁷Fe and ¹¹⁹Sn conversion electron Mössbauer spectrometry (CEMS).

Thin films of Sn_{1-x}⁵⁷Fe_xO₂ were implanted at room temperature with 1×10^{17} Fe ions/cm² and at 300°C with 5×10^{16} and 1×10^{17} Fe ions/cm². In both case, the acceleration energy was 100 keV. From TRIM calculations of implantation conditions of 1×10^{17} Fe ions/cm² we expected the iron profiles peaked at about 40 nm with a maximum Fe concentration of about 10 at. %. Only 5×10^{16} ions/cm² of the Mössbauer isotope ⁵⁷Fe were implanted while the balance was implanted with ⁵⁶Fe ions in the case of the 1×10^{17} ions/cm² implantation.

The implanted Fe ions exist as Fe(II) and Fe(III) in SnO₂ films, which partially also are reduced into Sn(II) on the implanted surface layer. The as prepared sample at room temperature and post-annealed samples did not show Kerr

effect, but the sample implanted with 1×10^{17} Fe ions/cm², while heated at 300°C, showed Kerr effect although the magnetic sextets were not observed in ⁵⁷Fe CEM spectra. The Kerr effect disappeared after post-annealing. It suggests that the number of magnetic defects decreases by absorption of oxygen.

For 5×10^{16} ⁵⁷Fe ion implanted samples, magnetic components with broad peaks were observed, but not for 1×10^{17} Fe ion implanted sample. This suggests that diluted Fe ions may induce the dilute anti-ferromagnetism or the paramagnetic relaxation because the Kerr effect was not clearly observed.

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3P01 Origin identification of building stone using prompt gamma-ray analysis

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A lot of cheap foreign building stones compared with domestic production are used in Japan. The quality evaluation is chiefly performed by the naked eye observation, and the method of a scientific evaluation has not established it. Elements in stones are estimated to reflect elemental features of producing districts. The origin identification method of building stone using a prompt gamma-ray analysis (PGA) was examined. PGA was performed at JRR-3 installed at JAEA. Eleven elements, H, B, Na, Al, Si, K, Ca, Ti, Fe, Sm and Gd were measured by PGA.

3P02 Analysis of metal concentration in aquatic insect larvae by INAA

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Elemental concentrations of aquatic insect larvae and attached algae in uncontaminated river were analyzed by INAA with k_0 -standardization method. The aquatic insect larvae found were all intolerant species. No significant difference was observed on the elemental concentrations of aquatic insect larvae and the attached algae along the river. Similar elemental concentrations were observed on the aquatic insect larvae collected at a fixed sampling point for two years. An analysis by the ratio matching technique indicated a higher generic relationship between aquatic insect larvae and attached algae than river water.

3P03 Instrumental neutron activation analysis of extractable organohalogens (EOX) in zooplankton and fish samples collected from the northern North Pacific Ocean

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Conservation, Ehime University)

Organohalogen compounds such as DDTs, PCBs and dioxin related compounds are persistent and ubiquitous in the environment, and accumulative and toxic for organisms. Firstly, these compounds are concentrated in zooplankton and fish in aquatic environment, secondly, are biomagnified among food web animals as fish-eating organisms, namely marine mammals and seabirds. Consequently, it is important to determine the levels and accumulative properties of halogenated organic compounds in zooplankton and fish samples in order to understand the environmental behavior of the compounds. The present study was undertaken to investigate the concentrations and residue profiles in zooplankton and fish samples collected in the northern North Pacific Ocean.

The detectable levels of EOX were presented in the samples: 0.765-1.68 for extractable organochlorine (EOCl), 0.096-0.128 for extractable organobromine (EOBr) and 0.0301-0.0862 for extractable organoiodine (EOI). The concentration order was EOCl > EOBr > EOI. The concentration ratios (fish / zooplankton) for each halogen were 0.93 (average) and 0.61 (min.) to 1.3 (max.) for EOCl, 0.86 (0.76 – 1.0) for EOBr and 0.89 (0.43 – 1.2) for EOI. The results of detail investigation show the presence of bio-accumulative organic chlorinated compounds in the northern North Pacific marine environment.

3P04 Separation of actinides(III) from lanthanides(III) by extraction chromatography using

N,N'-dialkyl-*N,N'*-diphenylpyridine-2,6-dicarboxyamides

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Separation of actinide(III) such as americium and curium with long-term α -radiotoxicity from high level radioactive waste containing lanthanide(III) is indispensable to attain transmutation of actinide(III) successfully. Extraction chromatography is one of the promising separation techniques for recovering small amounts of target components from a solution. In this study, to attain the separation of actinides(III) from lanthanides(III) by extraction chromatography, *N,N'*-dialkyl-*N,N'*-diphenylpyridine-2,6-dicarboxyamides (PDA) was synthesized as the extractants. This extractant system is prospective to selectively recognize actinides(III) over lanthanides(III) since this has nitrogen donor (pyridyl group) between two amidic functions. The extraction chromatography resin was prepared by contacting XAD4 (amberlite) with a methanol solution of PDA. The distribution coefficient, K_d , of M(III) to the resin was measured by batch method. The K_d of both Am(III) and Eu(III) increased with an increase of HNO₃ concentration in the range of 2 – 5 M. The separation factor (= $K_d(\text{Am})/K_d(\text{Eu})$) at 3 M HNO₃ was determined to be 6.9. From the result of column experiment, it was found that a complete separation between Am(III) and Eu(III) was archived.

Present study includes the result of “Development of separation technology of transuranium elements and fission products by using new extractants and adsorbents” entrusted to Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

3P05 Back Donation Properties of 5f of Pu and Cm to Organic Ligands from the Standpoint of Partial Density of States

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(JAEA,¹ LBNL²)

It is considered that the 5f orbitals of actinides show more itinerate properties than the 4f ones of lanthanides and however, these shielded by closed 6s and 6p orbitals could not participated in chemical bonding with molecular materials like organic ligands. However, it has been pointed out that there are significant differences on the interaction between soft donor ligands and actinides. In this presentation, the chemical bonding properties between actinides (Pu, Cm) and ligands and back donation properties of 5f electrons to the ligands on the basis of experimental partial density of states (PDOS) will be talked.

3P06 Application of imaging plate to measurement of size distribution of radioactive nano particles using Graded Screen Array

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Graded Screen Array (GSA) is often used in order to measure a size distribution of radioactive aerosols which are generated in an accelerator room. In this study, we applied imaging plate (IP) to measurement of radioactive aerosols using GSA so that experiments become facile and rapid. Irradiation was carried out at 46MeV electron linear accelerator in Reactor Research Institute, Kyoto University. We placed a flexible tube made of stainless steel behind a Ta target, and introduced the aerosol-free air into the tube at the rate of 5.7L/min. We collected radioactive aerosols by GSA for 20min during irradiation. The GSA is composed of five wire screens which have different mesh size and a PTFE filter. IP was exposed for 20min. From the detected values of IP we calculated penetration efficiencies for screens and obtained a particle size distribution by fitting a penetration efficiencies curve. It was found that geometric median diameter and geometric standard deviation were 28-31nm and 2.3-2.8, respectively. It was shown that this system can be applied to measurement of size distribution of radioactive aerosols in the accelerator room.

3P07 X-ray powder diffraction method for determination of trace asbestos in building materials by SAGA - LS

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Asbestos is widely known to be highly toxic. The inhalation of asbestos fibers can cause serious illnesses, including mesothelioma and asbestosis. Since the mid 1970s, many uses of asbestos have been banned in Japan. According to the Japanese law, it is necessary to analyze the asbestos concentration in the building materials in case of the dismantlement. However, it is not easy due to the trace level. In this investigation, we studied the X-ray powder diffraction method for determination of trace asbestos in building materials by synchrotron radiation at Kyushu synchrotron light research center.

3P08 Recent Environmental Tritium Levels at Kyushu Island, Japan

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Recent environmental tritium concentration shows fairly low level, because a large amount of the tritium released into the atmosphere in early 1960's by nuclear test has almost decreased. Also the nuclear power plants are another source of tritium. Two PWRs are under operation at Kagoshima Prefecture. We present the recent tritium levels of atmospheric HTO, seawater and precipitation.

Atmospheric HTO were continuously collected each month at three sampling sites. In this study, the results mean average concentration of tritium for one month. The tritium levels of HTO at the site close to PWRs showed slightly higher than other sites and they are almost same as 1980's atmospheric HTO. Seawater samples were also collected at three sampling sites. Tritium levels in seawater close to PWRs were almost same as control sites, but sometimes showed high levels presumed to be influence of PWRs. The tritium levels without influence of PWRs were under 15% of 1980's seawater. Each precipitation was collected at Fukuoka City. The difference of tritium levels with each precipitation was for the origin of air mass.

3P09 Investigation of extracting method of iodine for ¹²⁹I/¹²⁷I in natural water samples

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The extracting method of I isotopes in brine sample with ¹²⁹I/¹²⁷I of 10⁻¹³ of order of magnitude was investigated. Isotopic ratio of ¹²⁹I/¹²⁷I in iodine reagent was compared with different purity of NaCl reagent, 99.5 % (Wako Co. Ltd.) and 99.999% (Aldrich Co. Ltd.). The 99.5 % of purity for NaCl reagent can be used to obtain ¹²⁹I/¹²⁷I ratio of 10⁻¹³ of order of magnitude in the iodine reagent. It was confirm that isotopic ratio of ¹²⁹I/¹²⁷I ratio in the iodine reagent and the brine with ultra low ¹²⁹I/¹²⁷I ratio (10⁻¹³) can be exactly obtained by some reagents. Though the iodine isotopes in the iodine reagent and the brine was separated by an anion exchange resin to obtained iodine sample water, the ratio of ¹²⁹I/¹²⁷I in the sample water was 10 times higher than that in the iodine reagent and the brine sample. The conditioning method for the anion exchange resin reported literatures was difficult to eliminate ultra small amount of ¹²⁹I and ¹²⁷I in the anion exchange resin, suggesting that the separating method of the anion exchange resin was not preferable for brine sample with 10⁻¹³ of ultra low ¹²⁹I/¹²⁷I ratio.

3P10 Calculation of ²²²Rn emanation fraction for soil of single grain structure by Monte Carlo simulation

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In order to predict the radon emanation fraction of soil, we developed a soil model and calculated the radon emanation fraction of soil by Monte Carlo simulation. In this model, soil grains are packed in the simple cubic structure and the radon emanation fraction was calculated as a function of grain size, moisture content and radium distribution. As a result, the radon emanation fraction greatly increased with increasing the grain size in the range of 10-100μm and then was saturated when the moisture content was 0%. As the moisture content became

higher, the radon emanation fraction increased and then was saturated at smaller grain size. Water in pore enhanced the radon emanation fraction because it plays an important role to stop radon atoms before embedded into adjacent grains. On the other hand, the validity of the present model was evaluated in comparison with some experimental data. Since the model calculations were comparable with the experimental data, the present model would be more practical and available to estimate the radon emanation fraction of soil.

3P11 Chemical compounds and radioactivities in rocks sampled around radon hot springs

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The hot spring at Misasa in Japan and the gallery at Badgastein in Austria are world-famous for radon therapy. However, the mechanism has not yet been understood because the internal movement of inhaled radon is not obvious. In this study, for the first step to elucidate the mechanism of the radon therapy, the chemical compounds of rocks (grain size 250–500 μm) sampled at Misasa and Badgastein were identified by inductively coupled plasma-atomic emission spectrometry and X-ray diffraction. The radium activities and the radon emanation fractions were then measured using a high-purity germanium detector and a scintillation cell. As a result, the Misasa rock was probably composed of quartz, albite and microcline. The Badgastein rock was probably composed of quartz and muscovite. The radium activity of the Misasa rock (895 Bq/kg) was much lower than that of the Badgastein rock (7064 Bq/kg). In contrast, the radon emanation fraction of the Misasa rock (28.5%) was much higher than that of the Badgastein rock (2.1%). This may be caused by the difference in radium distribution inside grains. It is known that radium distribution is one of factors that contribute to the radon emanation fraction. Radium atoms may localize around the surfaces of grains of the Misasa rock that was weathered, although they may exist uniformly in grains of the Badgastein rock that was fresh. This difference possibly reflects the result of the radon emanation fraction.

3P12 The most suitable measurement of carbon -14 in bioethanols by liquid scintillation counters

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Toward the low carbon society, production of bioethanols prosperous. The bioethanol and ethanol obtained by chemical synthesis of oil are distinguished by measurement of ¹⁴C in ethanol with the liquid scintillation counter (LSC). Radioactivity of ¹⁴C in marketed ethanols were measured by using "OptiPhase HiSafe 2" and "Ultima Gold F" scintillators. The radioactivity of ¹⁴C in ethanols produced by the fermentation method and obtained by the chemical synthesis of oil is 14.05 - 14.66 dpm/g · C and 0.64 - 0.70 dpm/g · C respectively. Ethanol by chemical synthesis of oil included a very small amount of ¹⁴C. For this cause, there are thought that pollution with modern carbon in chemical synthesis and a background sample not being reasonable.

3P13 Measurement of atmospheric krypton-85 concentration

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One of the most important man-made radionuclides in the environment is krypton-85 (Kr-85). This nuclide has been released from nuclear fuel reprocessing plants in Europe and from the facility recently started to operate at Rokkasho, Japan. To analyze atmospheric Kr-85 concentration, we constructed an apparatus which collect and separate Kr from other atmospheric component and prepare Kr-85 counting sample. The apparatus consist in dust filter, molecular sieve trap, charcoal trap and gaschromatography. Activity of Kr-85 is measured with a low-back ground liquid scintillation counter using quart counting vial. Total error of less than 3% associated with Kr-85 determination is achieved. The atmospheric Kr-85 concentration of $1.53 \pm 0.04 \text{ Bq/m}^3$ was obtained in 2008 at Fukuoka, Japan, which is within the range reported for atmospheric Kr-85 concentration of other places.

3P14 Studies on secular variation of radiocarbon contents in tree rings (6-8th century) of Yaku cedar by AMS.

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Atmospheric radiocarbon (¹⁴C) is produced in the stratosphere by cosmic ray reaction with nitrogen. ¹⁴C is absorbed photosynthetically by plants. Since the variation of cosmic ray intensity is influenced by solar magnetic activity, the production rate of the ¹⁴C should have an inverse correlation with solar activity. In order to know the secular variation of ¹⁴C concentrations in the atmosphere, we have determined ¹⁴C/¹²C ratios in a Yaku cedar (1632 year-old). Selected annual tree ring samples during 560-800 A.D. were used. In this period, there is very little data about secular variation of ¹⁴C measured for Japanese samples. In the analysis, we have developed an apparatus to make graphite targets for Accelerator Mass Spectrometry (AMS). Using a small sample size, we could measure ¹⁴C/¹²C ratios with reasonable precision and accuracy by AMS at MALT, University of Tokyo.

Results obtained in this study agreed with a ¹⁴C pattern reported by IntCal04 [Reimer et al. (2004)]. However, if we see details, our data showed a wide range and a short term trend of the variation. This is because resolution of our data (single tree ring measurement) is higher than those of IntCal04 in which tree rings of ten years were combined. Around 700 A.D., $\Delta^{14}\text{C}$ maximum is observed. It is interesting to note that there was a cold period around this age, suggesting possible relationships between the solar activity and the climate.

3P15 The concentration of cosmogenic nuclide in Tamagawa hot spring

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The concentration of ^{10}Be ($T_{1/2} = 1.5 \times 10^6$ y) and ^{36}Cl ($T_{1/2} = 3.0 \times 10^5$ y) in Tamagawa hot spring water were investigated during 2003 to 2007. All samples were measured by accelerator mass spectrometry (AMS) at MALT, the University of Tokyo. The ^{10}Be concentrations in Tamagawa hot spring water were $(3.6 \pm 0.4) \times 10^7$ atoms kg^{-1} , which are 7 times higher than rainwater. The ^{36}Cl concentrations were not determined, since $^{36}\text{Cl}/\text{Cl}$ were below the detection limit of the MALT. The low atomic ratio of $^{36}\text{Cl}/\text{Cl}$ was attributed to volcanic gasses with high Cl content. In addition to AMS measurements, stable Be isotope concentrations were measured by ICP-AES. The ^9Be concentrations in Tamagawa hot spring water were 2.7 ± 0.5 ppb and the atomic ratio of $^{10}\text{Be}/^9\text{Be}$ were $(2.1 \pm 0.4) \times 10^{-10}$. Tamagawa hot spring water generated from rainwater in underground by adding volcanic gasses, so it was considered that the Be isotopes in Tamagawa hot spring water were leached from soil and volcanic rock. The $^{10}\text{Be}/^9\text{Be}$ ratio of Tamagawa hot spring water ranged between the ratios of soil (10^{-8}) and volcanic rock (10^{-11}). Assuming simple mixing of those sources, the Be isotopes in Tamagawa hot spring was contributed over 98% by volcanic rocks.

3P16 Environmental radioactivity measurement by simple handmade PHA module for understanding of measurement procedure

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For the purpose of the understanding on the measurement procedure of radioactivity, simple PHA module was made, and it was applied to the environmental radioactivity measurement. The assembly of the module was PIC microcontroller contained 10bit ADC and some circuits, RS232C format data was output in list mode for pulse input. Though the accuracy is low, it has the sufficient performance for the detector of the low resolution such as NaI(Tl) detector. And, it is possible to record the data input time by using timer of PC in tens millisecond unit, it is applicable for time interval analysis such as ^{220}Rn and ^{222}Rn measurement.

3P17 Study on the reversed phase chromatography in trace concentration for solution chemistry of rutherfordium.

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In order to research about the chlorocomplexation of rutherfordium, we measured distribution ratios (D) of its homologues, Zr and Hf with both of the batch method and on-line experiment with micro columns in reversed phase chromatography. In the experiments, we utilize the resin containing triisooctylamine as extraction solvent. The batch experiment was performed by using the tracers of ^{88}Zr and ^{175}Hf . The on-line experiment was performed by using $^{89\text{m}}\text{Zr}$. The results of the batch method show that the measured D values of Zr are larger than those of Hf in 7.5-10M HCl. Measured D values with on-line experiment don't agree with those by the batch method. It is considered that reaction for the on-line experiment doesn't come to equilibrium. We need to investigate the rate-determining step in the reaction process for the on-line experiment.

3P18 Preparation of a rotating ^{248}Cm target of GARIS

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We plan to study chemical properties of superheavy elements by using a gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator (GARIS). In this work, we studied a procedure to prepare a large and uniform Cm_2O_3 target by a molecular plating method for irradiations using a rotating target system of GARIS. Prior to the ^{248}Cm target, the electrodeposition conditions were optimized by preparing ^{147}Sm targets. Six microliters of 0.1 M HNO_3 containing 700 μg of ^{147}Sm were mixed with 5.5 mL of 2-propanol, and the mixture was filled into the electrodeposition cell. The target made in this study is banana-shaped with the active target area of 2.04 cm^2 . At the applied voltage of 1000 V, the deposition yields increase with increasing the time for electrodeposition from 52% at 2.5 min to 100% at > 7.5 min. The uniform target layer is visually observed, and the average-target thickness is 350 ± 10 $\mu\text{g cm}^{-2}$ for the targets electrodeposited for ≥ 7.5 min. The deposition yields also increase with increasing the applied voltage from 30% at 200 V to 100% at > 600 V during the 10-min electrodeposition.

The 8 pieces of the target made in this study will be arranged on the rotating wheel of 100 mm diameter. In the meeting, we will also report the preparation of the rotating ^{248}Cm target and its test irradiation with a high-intensity ^{18}O beam.

3P19 The liquid-liquid extraction behavior of Fm produced by the $^{238}\text{U}(^{16}\text{O},4n)^{250}\text{Fm}$ reaction

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The extraction behavior of fermium (Fm) has been investigated as part of the liquid-liquid extraction experiment of actinoid elements and superheavy elements. Fm was produced by the $^{238}\text{U}(^{16}\text{O},4n)^{250}\text{Fm}$ reaction using the RCNP AVF cyclotron. Reaction products recoiling out of the target were transported to a chemistry laboratory by a He/KCl gas-jet transport system. Transported products were dissolved in 100 μL of hydrochloric acid. Equal volume of 0.1 M thenoyltrifluoroacetone-carbon tetrachloride solution was mixed with aqueous solution and the mixture was shaken for 20 min. The distribution ratio was obtained from the radioactivities of the organic and aqueous phases measured by a α -ray spectrometry. To compare the result, lanthanides terbium (Tb), dysprosium (Dy), and erbium (Er) isotopes were produced by the cyclotron and distribution ratios were obtained by the similar way. The distribution ratios of other lanthanoids were also investigated by using RIKEN multitracers.

The distribution ratios of lanthanoids increase with decreasing the ion radius and with increasing pH. Although the obtained data for Fm suggests that Fm has different trend from lanthanoids, conformation of the result requires additional measurements because the counts of Fm were very few in this experiment.

3P20 Study on the synthesis of heavy elements through nuclear fusion induced by heavy ion on rare earth target.

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We performed an experiment for the synthesis of heavy elements with oxygen-16 and a rare earth nucleus of thulium-169, the latter of which is a deformed nucleus like many nuclei of actinides. The formation cross sections of evaporation residues through the nuclear fusion process were measured by using a stack technique and a gas-jet technique in the energy range of 72 to 116 MeV. The cross sections derived from the radioactivities of the products were compared with the theoretical values calculated by ALICE code. As a result, some notable differences between the experimental values and the theoretical values were found in the iridium products.

3P21 Application of laser ionization mass spectroscopy for gas phase chemistry

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In order to study chemical properties of transactinid elements it is necessary to apply rapid chemistry such as gas phase chemistry. Gas phase chemistry is the method of studying volatility of complexes of the objective elements. In our group, the volatile dipivaloylmethane (DPM) complex have been studied. However, their chemical forms have not been made clear yet. Therefore, the mass spectrometer has been used for elucidation of their chemical forms. There are two conditions of ionization for mass spectroscopy. First condition is that the ionization has nondestructivity, and the second is high-selectivity. As the ionization method to satisfy those conditions, Resonance Enhanced Multi-Photon Ionization (REMPI) was adopted. In the previous experiments of using an ion chamber, the result that DPM was ionized with REMPI method was observed. In the experiments of the mass analysis with REMPI at the resonance wavelength, $m/z=57$ and 127 (these are the fragment ions of DPM) were strongly observed and the ratio of strength of $m/z=57$ to that of 127 was different from the result of the EI method. The molecular ion of DPM, $m/z=184$, was weakly observed. Moreover, measurement of those ions at various wavelengths was performed.

3P22 cancelled

3P23 Studies on volatile behavior of group-4 elements for the gas phase chemistry of Rf

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Gas phase chemistry has been applied to the chemistry of superheavy elements because it allows separating them continuously and rapidly. In order to investigate of the chemical property of Rf, the volatile property of Zr which is lighter homologue has been studied in our group. Experiments of the gas phase chemistry of Zr and Hf were performed at JAEA and RIKEN. The nuclear reaction products attached to carbon clusters were introduced into the reaction section kept at

a high temperature. The volatile species produced by reacting with HCl in the reaction section passed through the isothermal column, and were deposited on the cooling section. After optimizing the experimental condition, for example the reaction temperature and the flow rate of HCl, the growth and decay of the activity of the volatile products was measured. Finally, the isothermal chromatogram for Zr was constructed. The adsorption temperature of the volatile compound of Zr was relatively high, and this result was similar to the previous report of our group.

3P24 Detection for alpha-particles of short-lived francium with flow-type liquid-scintillation counter

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A flow-type liquid-scintillation counter (LSC) was newly designed for detection of α -particles from heavy elements. Although a LSC has high detection efficiency but poor energy resolution, reduction of background is required. A new method for decreasing β -background based on difference in the range between α and β -particles was developed by using extra fine Teflon tube. Furthermore, as a test experiments for detection of short-lived heavy elements, α -particles of ²¹³Fr ($T_{1/2} = 34.6$ s) were detected with the flow-type LSC using extra fine Teflon tube. ²¹³Fr was produced by ²⁰⁹Bi(¹⁶O, 4n) reaction using the AVF cyclotron at the Research Center for Nuclear Physics (RCNP). The reaction products were transported to the chemistry laboratory by KCl/He gas-jet and collected 1 minute on a dissolution apparatus. The collected isotopes are dissolved with 25 μ L of water, mixed with 100 μ L of emulsifying scintillator, flowed into i.d. 0.5 mm Teflon tube. The α -particles of ²¹³Fr were successfully detected under reduced background for high energy β -particles.

3P25 Precision measurement of the half-life of Nb-90m using a gas-jet transport system

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In order to precisely determine the short half-life of ^{90m}Nb, we have developed a gas-jet transport system on the beam line of the RIKEN K70 AVF cyclotron. In this study, we determined the half-life of ^{90m}Nb in several of chemical states. Niobium-90m was produced in the nuclear reaction ^{nat}Zr(p, xn)^{90m}Nb. The reaction products recoiling out from the target were stopped in the helium gas, attached to KCl aerosol particles, and transported onto a polyester film. They were dissolved in 50 μ L of 20 M HF. The solution was placed in a polyethylene tube and subjected to γ -ray spectrometry with a HP-Ge semiconductor detector. The half-lives of ^{90m}Nb were determined by the reference source method using ¹³⁷Cs as a reference source. To obtain good statistics, the above procedures were repeated 120 times. As a result, the half-life of ^{90m}Nb in 20 M HF was found to be 18.97 ± 0.03 s, which is consistent with the previous value reported by Smend *et al.* The other half-lives obtained will be reported at the presentation.

3P26 Studies of extraction behavior of tungsten for chemical studies of seaborgium (element 106)

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The extraction behavior of tungsten (W) under extremely low concentration was investigated for the chemical studies of seaborgium (element 106). Tungsten isotopes, ¹⁷³W ($T_{1/2} = 7.6$ min) and ¹⁷⁴W ($T_{1/2} = 31$ min) were produced in the ^{nat}Dy(¹⁶O,xn)¹⁷⁴W and ^{nat}Gd(²²Ne,xn)¹⁷³W reactions, respectively, using RCNP and RIKEN AVF cyclotrons. Reaction products recoiling out of the target were transported to a chemistry laboratory by a He/KCl gas-jet transport system. Transported products were dissolved in 200 μ L of hydrochloric acid. Equal volume of 0.05 M tetraphenylarsonium chloride-chloroform or 0.05 M Aliquat 336-chloroform solution was mixed with aqueous solution and the mixture was shaken for 3 min. The distribution ratio was obtained from the radioactivities of these two phases measured by γ -ray spectrometry. The distribution ratio was very low ($< 10^{-2}$) at 0.1-4 M hydrochloric acid and rised steeply at 4-8 M hydrochloric acid, indicating the formation of chloride complexes of W.

3P27 Influence of chrome on the formation of β -FeOOH in aqueous solution analyzed by Moessbauer spectroscopy

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Ferric oxyhydroxides (FeOOH) are main components of corrosion products of iron. Among the ferric oxyhydroxides β -FeOOH, which contains chloride ions in its structure, is considered to be formed in the coastal area. Although Cr is often added to steel in order to protect corrosion, influence of Cr on the formation of β -FeOOH is not well understood.

We have synthesized β -FeOOH from 0.1M FeCl₃ solution with Cr³⁺ by hydrolysis and analyzed it by Moessbauer spectroscopy and X-ray absorption fine structure (XAFS). As a result, three different Fe³⁺ sites were found in ⁵⁷Fe Moessbauer spectrum at 78K. Quadrupole splitting of the three Fe³⁺ components showed little changes. It implies that the symmetry of β -FeOOH was kept in spite of the addition of Cr. Moreover, radial structural functions (RSFs) of extended X-ray absorption fine structure (EXAFS) spectra on the Fe K-edge revealed that the length of Fe-O and Fe-Fe bond were not affected by the addition of Cr. These results showed that the fundamental structures of FeO₆ octahedral unit in β -FeOOH were not changed by the addition of Cr.

3P28 Spin States of Assembled Iron Complexes and Their Mixed Crystals

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We have studied spin-crossover behavior of assembled complexes. In particular, Fe (NCX)₂(bpp)₂ (X=S, Se, BH₃; bpp=1,3-bis(4-pyridyl)propane) has very rare structure which is 2D interpenetrated, and Fe(NCBH₃)₂(bpp)₂ shows spin-crossover behavior. In the present study, we performed mixed metals experiment to identify the character of assembled complexes. In a general way mixed metals experiment reduces Fe-Fe interaction but now, we pick on the difference of radius of ion, and we started mixed metals experiment of complex

Fe(NCX)₂(bpp)₂ with zinc or cobalt. (Ion radius of bivalent high-spin iron is 92pm, low-spin is 75pm, bivalent zinc is 86pm, bivalent high-spin cobalt is 88.5pm, low-spin is 79pm.)

We receive structure of complex Fe_{0.5}Zn_{0.5}(NCS)₂(bpp)₂ is equal to that of Fe (NCS)₂(bpp)₂ by powder X-ray crystallography. In M \ddot{o} ssbauer spectrum of mixed crystal, two independent doublets ascribed to Fe(II) HS state are observed, so that two iron sites are displaced unselectively. The rate of Fe(II) LS of Fe_{0.5}Co_{0.5}(NCSe)₂(bpp)₂ is larger than that of Fe (NCSe)₂(bpp)₂. Co is smaller than Fe, so Fe tries to become small (chemical pressure effect). A fastener effect of long alkyl chain is usually seen, but chemical pressure of mixed crystals is rare. This effect is observed in NCSe complex, but not in NCS complex. This is derivable from nature of Se which is softer than S.

3P29 Construction and functional expression of iron(II) spin-crossover metallomesogen

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Metallomesogen consists of transition-metal complex and long alkyl chains. Transition-metal complexes with the dynamic electronic states express the functionality by the spin state change. So the complexes with long alkyl chains have both the functionality by the spin state and liquid crystallinity. This time, we attempted the construction of metallomesogen with switching function by introducing long alkyl chains to the spin-crossover complex.

We synthesized the metallomesogen, [Fe(3Cn-bzimpy)₂](BF₄)₂ (n=8~22) newly. [Fe(3C16-bzimpy)₂](BF₄)₂ showed the transition from solid to B2 mesophase at 331K, the transition from B2 mesophase to SmA mesophase at 447K and the transition from SmA mesophase to liquid at 466K. And the relative permittivity of this compound rose at liquid crystal transition temperature, and in *P-E* measurement, the compound showed hysteresis behavior in B2 mesophase, thus showed ferroelectricity. In magnetic measurement, the compound shows spin-crossover phenomenon at liquid crystal transition temperature. In single-crystal X-ray structure analysis, it was found that six long alkyl chains of the compound were divided into two and four. We succeeded in the creation of ferroelectric liquid crystal in the molecular without chiral source and bent structure.

3P30 Redox control of technetium cluster by the face capping ligand

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Octahedral hexanuclear cluster complexes have interesting optical and redox properties. In this study, redox properties of two new hexatechnetium complexes with mixed sulfide and bromide as the capping ligand [Tc₆(μ_3 -S₇Br)Br₆]³⁻(**1**) and [Tc₆(μ_3 -S₆Br₂)Br₆]²⁻(**2**) are reported.

The complexes **1** and **2** were prepared by the high temperature reaction of technetium, sulfur, bromine and potassium bromide. These complexes were separated by column chromatography. The structures were determined by single crystal X-ray analysis. The Tc₆ octahedral cores are surrounded by two ligands, sulfide and bromide. Both of the complexes **1** and **2** have six bromides as terminal ligand.

The respective redox potentials of the **1** and **2** was measured by cyclic voltammetry in (Bu₄N)PF₆-CH₃CN. The reduction potential assignable to Tc^{III}Tc^{II}(25e)/ Tc^{III}₆(24e) process are observed at -1.0 V (**1**) and -0.32 V (**2**), respectively. The oxidation potentials assignable to Tc^{III}₆(24e)/ Tc^{III}₅Tc^{IV} (23e) process are not observed both for **1** and **2**. The reduction potential of **2** shifts 680 mV more positive than that of **1**. The hexatechnetium complexes become easier to reduce with increasing the number of capping bromide.

3P31 Electronic phase separation as a root of colossal magnetoresistance

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Modern data storage systems are dominantly based on the recording and reading of the magnetization of small magnetic domains. In order to convert the magnetic signal into electronic pulse, magnetoresistive (MR) materials are essential. The more sensitive the magnetoresistive material is, the higher the density of stored data and the faster the writing/reading process can be. In the previous decade, a new class of magnetoresistive materials were also discovered which show significantly higher MR around their magnetic ordering temperature. The advantage is a bulk property of several types of compounds that is so called colossal magnetoresistance (CMR).

Typical examples for CMR materials are the doped cobaltate perovskites, cation disordered double perovskites, or chalcogenide spinels. The most intriguing fact is that these materials differ in composition, crystal structure, and magnetism each other; still they all show the colossal magnetoresistance.

In order to elucidate the source of CMR effect, and so to be able to improve magnetoresistive sensitivity, finding a common feature of CMR materials is essential. In this review we present our recent studies of the local and bulk electronic/magnetic properties of several CMR materials. Investigating the effect of small modulations (doping with ions, increasing cation disorder), we were able to determine that the common point in the aforementioned CMR materials is a nanoscale electronic phase separation, which seems to be responsible for the observed magnetoresistance.