English Abstracts

1S01: Results of the Hayabusa mission and goal of preliminary examination of the samples.

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The Hayabusa spacecraft arrived at the S-type asteroid 25143 Itokawa in November 2006, and succeeded to reveal astounding features of the small asteroid (535 × 294 × 209 m) [1]. Although the sample collection did not made by normal operations, it is considered that a small amount of samples, at least a few particles of approximately ten to a few hundred micron meters in size, were collected. The sample capsule was successfully recovered on the earth in June, 2010. After picking up of sample particles from the sample container, some of them will be examined preliminarily within one year in prior to detailed analysis phase. The following scientific achievements will be expected; (1) characterization of the surface material of Itokawa (classification: ordinary LL chondrite or primitive achondrite, formation age, formation process and conditions, density, etc), (2) processes on an ancestor boby of Itokawa and accretion into Itokawa (brecciation, degree of impact, etc.), (3) interaction with space environment (proof of space weathering, isotopic compositions of oxygen and noble gases of the solar wind, age of regolith, gardening rate, etc.), and (4) materials fallen onto the surface (primitive materials, such as organic materials and/or differentiated granitic fragments).

[1] Fujiwara et al. (2006) Science, 312: 1330.

2S01: Chemistry of Superheavy Elements – Experimental Achievements and Perspectives

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Ground-breaking atom-at-a-time experiments with the lighter superheavy elements (SHE) Rf, Db and Sg showed properties typical for group 4, 5 and 6 elements, respectively. Recent chemical studies aimed at (i) unraveling detailed properties of Rf and Db in aqueous solution, (ii) determining properties of HsO_4 in the gas-phase, and (iii) obtaining first chemical information from the element 114 region. Common to all these experiments is - in addition to obtaining basic chemical knowledge - the quest for the influence of relativistic effects on chemical properties of SHE. In parallel to the advancement of experimental knowledge, progress in modern relativistic quantum theory has provided crucial guidance and understanding. Moreover, chemical separations were applied for nuclear studies in the region of deformed doubly-magic ^{270}Hs. The progress in SHE chemistry was closely linked to improvements, and to new and automated techniques, from target technology to transport systems, chemical separation devices, and innovative detection system. Most recently, coupling of chemical set-ups to recoil separators opens new fields in SHE chemistry and provides great perspectives for the future. Among those are not only experiments in the element 114 region but also those paving the way to the organometallic chemistry of transactinides.

2S02: Study on hot atom chemical process of energetic hydrogen isotopes in carbon-related materials

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Hydrogen isotope retention behaviors for carbon-related materials, SiC, graphite, WC and simultaneous C^+-D_2^+ implanted W for fusion reactor application were summarized. The deuterium retention for SiC and graphite (HOPG) was higher than that for WC and simultaneous C^+ implanted W. XPS spectra indicate that the large chemical shift was observed for SiC and graphite, but no peak shift was found for WC and C^+ implanted W, indicating that most of deuterium was retained in the intrinsic defects, interstitial sites and/or irradiation defects. However, in the case of SiC and graphite, deuterium does not remained in the interstitial site, but bound to Si and/or C due to high affinity with hydrogen isotope. In the case of simultaneous irradiation, the deuterium trapping is dynamically proceeded with the competition between the dissociation of matrix and chemical trapping of deuterium with constituent atoms.

2S03: Study on synthesis and properties of technetium complexes

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Among the various technetium complexes, the chemistry of technetium-technetium bonded complexes composed of three or more nuclei is still limited. In this study, the synthesis, structures, and physical properties, and electronic structures of the various octahedral hexatechnetium complexes and several mononuclear technetium complexes were investigated.

Discrete and polymeric chalcogenide-capped octahedral hexatechnetium(III) complexes were synthesized and structurally characterized. The Tc-Tc distances in the Tc_6Q_8 core are longer with the order of Q = S < Se < Te. Discrete sulfide- and selenide-capped hexatechnetium(III) clusters were synthesized by the reaction of a discrete molecule or a polymeric complex with excess cyanide. The series of the chalcobromide-capped hexatechnetium complexes were synthesized and characterized.

The redox potentials of the Tc_6(23e/24e) process in the technetium complexes were more positive than that of the Re_6(23e/24e) in the hexarhenium analog. On the other hand, the potential of Tc_6(22e/23e) shifts to negative as compared with the potential of Re_6(22e/23e), which is the opposite of the tendency usually observed in technetium and rhenium complexes.

The HOMO/LUMO levels in $[Tc_6Q_8(CN)_6]^{4-}$ (Q = S or Se) were determined. The energy level of HOMO is lowered compared with those of the rhenium congeners. The HOMO-LUMO energy gap of the hexatechnetium complexes is smaller than that of $[Re_6Q_8(CN)_6]^{4-}$.

The seven-coordinate technetium(IV) and technetium(V) complexes were synthesized and their properties were investigated.

3S01: New physiological activity of plants revealed by application of radiation and radioisotopes.

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Since plants live on only 17 inorganic elements and water, the application of radiation and radioisotopes opens an important field to analyze living plant activity.

Though water plays an important role for plant physiology, the behavior of water itself has not been known well, mainly because of lacking tools. We applied neutron beam analysis to image water profile within a plant, not only above-ground part but also the roots imbedded in soil. Morphological development of the root as well as the water uptake manner of the roots in soil was visualized by constructing spatial image of the root.

To analyze water movement itself, we employed positron emitters, ^{18}F and ^{15}O. In the case of ^{15}O, whose half-life is only 2 min., ^{15}O-water was supplied to a soybean root and the increase of radioactivity in the stem was measured. To our great surprise, we found that tremendous amount of water was always flowing out from xylem cells, which was regarded as mere pipe to transport water or nutrient. The water coming out from xylem cells was pushing the water already existed to re-enter into the xylem tissue. That is, what we found was water circulation in a plant! It was simulated that within 20 min., about half of the water already existed in the stem was replaced with the fresh water coming up from the root.

In the case of nutrient elements, first of all, we analyzed thousands of tissue samples of plants along with the developmental stage, by an activation analysis. There was an element specific pattern of distribution profile within a plant and the features of the profile were not changed throughout the life cycle. The plants growing in nature showed specific accumulation of the elements which reflected the nature of the soils, thus we could separate the plants according to the production sites.

To analyze the nutrient uptake manner in a living plant, we developed two types of real-time isotope imaging systems. One was to image whole plant and the other was to image microscopic tissue, remodeling a fluorescent microscope. Using these systems, we were able to show how roots accumulate ions in culture solution and transfer to the above-ground part. In the case of rice, ^{32}P-phosphate uptake manner was compared between water culture and soil culture. The amount of ^{2}P-phosphate taken up from water culture was very high, more than 100 times, compared to that of soil culture.

Nowadays, fluorescent imaging is widely used but for plant research, radioisotope imaging has two great advantages. One is that it allows imaging under light and the other was it enables to handle the numerical treatment of the image, for the image was based on radioactivity.

K101: Role of neutron activation analysis in trace element analytical methods

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The k_0-standardization method of instrumental neutron activation analysis (INAA) has been evaluated by determining the reference materials associated with air pollution, NIST SRM 1633a (Coal Fly Ash), NIST SRM 2783 (Air Particulate on Filter Media), and NIES CRM No.8 (Vehicle Exhaust Particulates). The analytical results of NIST SRM 1633a obtained by different facilities of the reactors were consistent each other, and the deviation from certified (reference) values for 17 elements were within 3%. It was ascertained that the present k_0 INAA is very accurate. This method was applied to the aerosol samples taken in Ibaraki prefecture. Trace element contents in particulate matters with different sizes were determined, and the distribution of elements and the regional variation were revealed. Nondestructive multi-element INAA is recognized to be still one of the most useful and important analytical techniques in trace element analysis.

K102: Importance of radiochemical neutron activation analysis – in case of the determination of ultra-trace rare earth elements in rock and meteorite samples

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Radiochemical neutron activation analysis (RNAA) involves radiochemical separation and purification of the target elements after the neutron irradiation of samples. As matrix elements including interfering elements can be removed, RNAA can determine ultra-trace elements. Here, we show the case of determining ultra-trace rare earth elements (REEs) in rocks and meteorites.

Geological standard rocks and Allende meteorite were subjected to RNAA in this study. The samples were irradiated with neutrons in the JRR-3 reactor of Japan Atomic Energy Agency. The RNAA procedure for the determination of REEs was mostly based on Minowa and Ebihara (2003) [1]. In addition to REEs, U concentrations were determined by using fissiogenic nuclides.

Ten REEs (La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb and Lu) were able to be determined. As ^{140}La, ^{141}Ce and ^{147}Nd are produced by neutron-induced fission reactions on ^{235}U in addition to neutron-capture reactions of individual target nuclides (^{139}La, ^{140}Ce and ^{146}Nd, respectively), their contributions were estimated and corrected for. Uranium was determined from the same samples as used for REEs by using fission products of ^{140}Ba and ^{140}La.

This study clearly indicates that RNAA can be the most suitable analytical method to determine very low concentrations of REEs in rock samples. We are confident that the importance of RNAA will not be changed even in future.

[1] H. Minowa and M. Ebihara, Anal. Chim. Acta 498 (2003) 25.

K103: The role of neutron activation analysis in the development of certified reference material

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In this talk, I would like to discuss the role of neutron activation analysis in the development of certified reference material. In 2007, neutron activation analysis recognized as primary ratio method by CCQM (consultative committee of amount of substance-metrology in chemistry). In addition, neutron activation analysis is well known as a non-destructive analytical method, which can determine most elements without any chemical pretreatments, that is, neutron activation analysis is basically free from a potential risk of loss and contamination during the sample preparation and measurement procedures. In particular, neutron activation analysis is very useful to determine mono-isotopic elements in environmental and biological samples, since the isotope dilution analysis that is one of the primary ratio methods cannot be applied to those elements. I will report the application of neutron activation analysis for the accurate determination of As in the arsenobetaine standard solutions.

K104: INAA attempting to get a fruitful national inventory map “Geochemical Map”

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Geochemical Map is the map representing the areal distributions of chemical elements and isotopes on the terrestrial earth. The map gives chemical information for environment and resource. Many elements for many samples must be analyzed to get a reliable geochemical map in wide area. XRF and ICP-MS are used for many researchers. I understand, however, INAA is the best techniques for the purpose. The INAA gives quite reliable data for Au, Ir, Os, As, Sb, Cr and REE. These are the important elements for environmental assessment and resource evaluation. The INAA provide a reliable data even for samples which contain hardly dissolvable minerals such as zircon. I will introduce our study in Aichi~Gifu area, and will demonstrate how the INAA is useful to draw the reliable geochemical map to cover Southeast Asia as well as Japan.

K105: Multi-element analysis of environmental samples by instrumental neutron activation analysis and multiple prompt gamma-ray analysis.

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Instrumental neutron activation analysis (INAA) is one of useful means for analyzing the distribution of multi-elements in environmental samples. We have applied this technique to examine the concentration and the vertical distribution of more than 30 elements in estuarine and tideland sediments by using with prompt gamma-ray analysis (PGA).

Sediment samples were collected at Yatsu tideland vertically. The sediment cores were approximately 50 cm long and then cut into 3-cm long pieces. Those pieces were put in oxygen-impenetrable plastic bags with N_2 gas immediately to prevent from air oxidation. We clarified the vertical distributions of elemental concentrations by INAA and PGA. As a result, big changes of them were found at *ca*. 25 cm from the surface in Yatsu tideland sediment core. It is thought that these changes were caused by neighboring reclamation. The vertical distributions of elemental concentrations were categorized into four groups according to the correlation matrix. By applying principal component analysis (PCA), it was suggested that sulfate ion was reduced under anaerobic environment and then various sulfides were formed in the sediments. It was supported by the result of Mössbauer measurement showing the vertical distribution of pyrite (FeS_2).

Recently, we have also applied multiple prompt gamma-ray analysis (MPGA) to pelagic sediments to examine the K/Ti ratio. The obtained ratios of sediments and standard reference materials have shown a good agreement with those by PGA.

K106: Applications of neutron activation analysis in environmental chemistry. –Presence of extractable organohalogens (EOX) in wildlife

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Many kinds and large amounts of synthetic organohalogen compounds are produced and are used in our daily life, and some of them introduced to the natural environment. In some cases, those substances that have dispersed in the natural environment are harmful for wildlife and for humans due to their persistency and their high toxicities. Neutron activation analysis (NAA) is suitable for trace amounts in biological samples such as wildlife in order to understand the levels as organic bound halogens, namely extractable organohalogens (EOX). The analytical procedures are as follows: extracting EOX with organic solvents from the sample, washing the extract with deionized and distilled water, fractionating with gel permeation column chromatography. Finally, NAA were performed with JRR-3 and JRR-4 of Japan Atomic Energy Agency located in Ibaraki, Japan. Gamma-ray spectrometry was performed just after irradiation. The gamma-rays from ^{38}Cl, ^{80}Br and ^{82}Br, and ^{128}I were measured by gamma-ray spectrometry. Gas chromatograph (GC)/ Mass spectrometry (MS) was performed with same samples.

K107: What can you know from trace elements in old iron?

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The use of iron started for about 4000 years in the history of mankind. Iron greatly develops human civilization, and it is still essential resource.

Iron is called steel or a cast iron by the concentration of carbon and characteristic property is different. In the impurity elements of iron, there is something for which information exists in the iron raw material has been left. Arsenic and antimony of the impurity are important elements for the identification of source of iron raw material. Namely, the As/ Sb concentration ratio is an index of source of iron raw material. An instrumental neutron activation analysis is one of the most suitable analytical methods for determination of Arsenic and Antimony in old iron as cultural properties. However, applications of the instrumental neutron activation analysis for cultural properties field in our country are not enough. The development of further uses to this field is expected.

K108: A value of multiple prompt gamma-ray analysis in an integrated analytical company

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Neutron activation analysis has been regarded to be one of the high sensitive elemental analysis methods as shown in text books. However, some of the nuclear reactors for research were stopped and doors of the other reactors are almost closed. Thus, utilization of the method in the industrial fields is opened only in the few nuclear facilities. In the circumstances, frequency in use of the method in the industrial fields is extremely going down. On the other hand, we know the rapid development of ICP-MS from 1988 makes this method to a pronominal of the ultra-trace elemental analysis method. Recently, Japan Atomic Energy Agency developed GEMINI（Prompt Gamma-Ray Analytical Instruments） and STELLA（Multiple Prompt Gamma-Ray Analytical Instruments） and promotes a utilization of Multiple Gamma-Ray analysis. Toray Research Center, Inc. cooperated to an acquisition of basic data for MPGA data base through the inter-organization atomic energy research program (2003~) or the promotion research with JST (2007~2010). In this presentation, based on the results of these research programs, a value of multiple prompt gamma-ray analysis in an integrated analytical company will be discussed.

Figure 1 shows 2-D spectrum of Cl 1000 μg added to NIST SRM 1632a (Bituminous Coal) obtained by using STELLA. Figure 2 shows 1-D spectrum of Cl obtained from the same measurement. In this case, determination limit of Cl was estimated to be 10 μg/g. For the other some elements, determination limits were ranged from 0.x – 10×x μg/g. This method is one of the ideal bulk analysis methods and the determination limit of Cl seems to be sufficiently useful in the industrial fields.

K109: Current request situation of contract activation analysis service in S.H.I. Examination & Inspection, Ltd. and trend in the future

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We have executed the contract Charged-Particle Activation Analysis (CPAA) by using our compact cyclotron for 20 years or more. CPAA is an analysis a light element in metallic materials and other materials. We have analyzed 3657 samples in these about 20 years.The requests of the customers have chiefly analyzed semiconductor materials in CPAA. In the first stage, this technique was used to develop the method of manufacturing semiconductor materials. The analysis of metallic materials increased after that, too. However, the number of requests has not increased in dramatic form.

And, we have been beginning the contract Neutron activation analysis by using the equipment of JRR-3 since 2008. We have executed the analysis that exceeds 3000 samples by using k0 method. The requests of the customers have chiefly analyzed environmental sample was analyzed in NAA.

It seems that the request amount in the future by 300-400 samples par year in our CPAA service. And, it is forecast that the number of requests doesn’t expand because our NAA service has the restriction at the machine time.

K110: Current Status of Activation Analysis Experiments in KURRI

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The operation of KUR with new low-enriched fuels has been started in May 2010, and neutron irradiation experiments have been also started in June 2010. For the purpose of upgrading the various systems for activation analysis experiments, improvements of facilities and installations of new equipments have been carried out in the down period of KUR. As improvements of facilities, control systems of pneumatic transport system, gamma-ray spectrometry systems including an auto sample changer, and other safe control systems have been developed for the advanced and convenient activation analysis experiments in safe environment. And as installations of new equipments, the prompt gamma-ray analysis equipment and the gamma-ray spectrometry system composed of a Ge-detector and a well-type NaI(Tl) detector for short-lived nuclei have been installed. The application range of neutron activation analysis method in KURRI is expected to spread by those improvements and installations. For the educational activity of activation analysis method, the practical training program intended for graduate students will start in 2010 for the purpose of attaining basic knowledge and technique of neutron activation analysis and radiochemical experiments.

K111: Development of MPGA in JRR-3 and J-PARC

TOH, Y.[1], FURUTAKA, K. [1], HARA, K. [1], HARADA, H. [1], KIMURA, A. [1], KIN, T. [1], KITATANI, F. [1], KOIZUMI, M. [1], NAKAMURA, S. [1], OSHIMA M. [1], MURAKAMI Y. [2]
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A neutron beam line and a detector system for MPGA were constructed at the neutron guide hall of JRR-3M at Japan Atomic Energy Agency (JAEA). The MPGA detector system consists of eight clover detectors with BGO Compton suppressors. Accidental coincidence rates increase with a square of an intensity of a neutron beam, while true coincidences increase only linearly with an intensity of a neutron beam. Therefore, a neutron attenuator system consisting of acrylic plates was installed in front of the MPGA beam line. Polymers, foods, and standard samples were measured using the MPGA detector system. Hydrogen backgrounds were reduced in gamma-ray spectrum of MPGA.

A pulsed neutron beam is produced by a pulsed proton beam in J-PARC. Neutron capture cross section shows sharp peaks, known as resonances, at energies specific for each isotope. The energy of a neutron can be determined by the TOF method where the flight time of neutrons is obtained by a time difference measurement. A signal-to-noise ratio in gamma-ray spectra of MPGA using TOF for energy selection will be more improved than that without TOF.

K112: Promotion of utilization for neutron activation analysis at JAEA

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In this lecture, activity of utilization promotion including neutron activation analysis that has stared as the common promotion project will be introduced while industrial promotion project of Prompt gamma-ray analysis by the trial-use program for neutron industrial applications will be introduced.

K113: Networking of neutron activation analysis people in Asia-Australia.

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Research reactors are essential for neutron activation analysis but their utilizations have been decreasing worldwide, especially in western European countries and US. Although Japan seems to have been following such countries, the current situation surrounding research reactors utilized for neutron activation analysis is rather stable, considering that the Kyoto University reactor and two reactors of JRR-3 and JRR-4 in Japan Atomic Energy Agency are currently running. However, these reactors were not operated from July 2009 to March 2010, which caused serious influence to us both in education and in research.

Nine Asian countries (Bangladesh, China, Indonesia, Japan, Korea, Malaysia, Philippines, Thailand and Vietnam) and Australia are allied under the frame of Forum for Nuclear Cooperation in Asia, which is financially supported by MEXT and the Cabinet office. Neutron activation analysis has been chosen as a project since the FNCA was established in 2000. In future, it is desired that the activity of the FNCA/NAA alliance will be extended so that unexpected suppression of reactors cannot be fatal in any FNCA countries.

K201: Radionuclides in Diagnostic Nuclear Medicine : Present and Future

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The present crisis of diagnostic nuclear medicine is a shortage of ^{99}Mo-^{99m}Tc supply from over-sea reactors. Several trials demonstrated a potential application of accelerator to produce abundant amount of ^{99m}Tc for medical use. Domestic production and stable supply is essential in future nuclear medicine.

In biomedical science, the target is a molecular basis of disease. ^{124}I is quite useful to label peptides, nucleic acids, and glycoprotein. The tracing of ^{52}Fe, ^{62}Zn, and ^{48}Ca may reveal new insight of physiology in immune system and intracellular signal transduction.

K202: Targeted radionuclide therapy – Current status and a perspective

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In Japan, 4 types of targeted radionuclide therapies (TRT) are currently available for routine use, including ^{131}I-sodium iodide for thyroid cancer and hyperthyroidism, ^{131}I-metaiodobenzylguanidine (MIBG) for malignant neuroendcrine tumors such as pheochromocytoma and neuroblastoma, ^{90}Y-labaled murine monoclonal IgG1 for B-cell lymphoma (radioimmunotherapy) (Zevalin®), and ^{89}Sr-strontium chloride (Metastron®) for palliation of painful bone metastases. Because of their targeting mechanisms, TRT is a true form of molecular targeted therapies.

As many of radionuclides emit γ ray or positron, *in vivo* distribution of radiopharmaceuticals can be delineated by scintigrphic technique with single photon computed tomography (SPECT) or positron emission tomography (PET), which makes TRT a surely unique therapeutic modality. That is, we can confirm targeting of radiopharmaceuticals in lesions. Furthermore, their distribution in normal tissues can be simultaneously monitored. In addition, we can obtain changes in their distribution with sequential image acquisition, which provides pharmacokinetic data on them. These features enable us predict therapeutic response of lesions and possible adverse reaction. Therefore, TRT is patient-specific, tailored treatment. Based on these concepts of TRT, we would like to propose to call TRT targeting confirmative therapy (TCT).

Therapeutic effects are currently provided with β particles of radionuclides. Cells within millimeter path length of β particles are irradiated, so that it is not necessary to target all cells in lesions. In addition, many of them are irradiated by several β rays (cross-fire effects), making the advantage of TRT over non-radioactive pharmaceuticals. Alfa emitters or Auger electron emitters with high LET (linear energy transfer) and RBE (relative biological effectiveness) are candidates for the next generation of therapeutic radionuclides.

What we have to consider in developing new therapeutic radiopharmaceuticals is a radiation safety issue. Non-gamma emitters or radionuclides with low energy, low-probability gamma emission would be better candidates for future targeted radionuclide therapy. For non-gamma emitters, we should need counterparts possessing gamma emission as a case of ^{90}Y-^{111}In in order to utilize the advantage of TCT.

K203: Application of RI Imaging in Pharmacological Sciences

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[Introduction] Radiotracer method using b^- emitters such as ^{3}H, ^{14}C, ^{32}P and ^{35}S has been widely applied in the field of pharmacological sciences because of its high sensitivity as compared to other methods. Recently, imaging with positron emission tomography (PET) becomes one of the key technologies in the drug developing process. In therapy assessment of new drug in animals and humans, PET imaging would be of great value to estimate pharmacokinetics parameters as well as pharmacological effects (including the adverse effects). The current state of application of PET imaging in the development of new drugs will be presented.

[Pharmacokinetics] Using ^{11}C- or other positron emitters-labeled new compounds, their pharmacokinetic studies can be performed in human subjects with PET imaging. This method is called as ‘micro-dosing test’, and absorption, distribution, metabolism and excretion in human can be directly measured by employing this micro-dosing method. The method seems to be of value as a screening test prior to starting clinical trials of the new compounds. Because of very short half-lives of positron emitters, the development of new radiosynthesis method is required.

[Drug-target interactions] Many compounds under developing are selective molecules to target proteins such as receptors and enzymes. Drug-target interactions in intact whole body either experimental animals or humans also can be measured by using selective PET radioligands. For example, the measurement of receptor occupancy, and rate constants (association or dissociation) of the compound for the target molecule could provide valuable information for preparing the clinical protocol. In order to apply this technology for the new target molecule, the new PET probe should be designed and evaluated, which is considerably difficult subject in future study.

[Pharmacological effects] The most important and valuable subject in PET imaging for development of new drugs is to find out the new biomarker, which enable us to evaluate pharmacological effects of the new compound in human especially in patients with target disease. The efficacy of the new drug can be obtained as changes in parameters of image.

The development of the new biomarker for PET imaging is useful for not only monitoring the new drug treatment, but also early diagnosis of disease as early as possible.

[Conclusion] PET imaging has high potentials as a key technology in developing new therapeutic drugs. The development of new imaging biomarker is the most important subject for the above purpose.

K204: Therapeutic effects for bone metastases using a-emitting radionuclides, ^{227}Th, ^{226}Th, ^{223}Ra in rats

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Due to short range and high liner energy transfer of alpha particle in human tissue, targeted alpha therapy has been promised for cancer therapy and many clinical studies have been performed. Especially, ^{223}Ra has been studied in a phase 3 as a therapeutic agent for bone metastases from prostate cancer. Radium-223 decays to stable ^{207}Pb with emission of about 28 MeV.

We have been studied another alpha-emitting nuclide ^{227}Th, which is the parent nuclide of ^{223}Ra. As the half-lives of ^{227}Th and ^{223}Ra are similar, it will be possible to avoid the high-dose alpha-particles of ^{223}Ra and its progeny nuclides during the initial blood circulation after administration of ^{227}Th. In addition, the radioactive growth of ^{223}Ra will prolong the effective irradiation duration. Thorium-227 emits alpha-particles with an average energy of 5.9 MeV.

Thorium-226 d is an alpha-emitting radionuclide which decays with a rapid cascade of 4 alpha particles with about 28 MeV of total energy. Because thorium is easy to label with EDTMP, ^{226}Th-EDTMP is also a promising agent for bone metastases.

In this study, we evaluated the therapeutic effects of three bone-seeking alpha-emitting radiopharmaceuticals, ^{223}RaCl$_2$, ^{227}Th-EDTMP, ^{226}Th-EDTMP from the point of view of the half-lives, and internal dosimetry.

K205: New Molecular Imaging approach using Electron Tracking Compton Gamma Ray with Possible Application of new various Radio Isotopes

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We have developed the Electron tracking Compton Camera (ETCC) with reconstructing the 3-D tracks of the scattered electron in Compton process for both gamma-ray astronomy and nuclear medicine. By measuring the direction of a scattered electron, the direction of the incident gamma ray is uniquely determined for an individual photon. Then ETCC can provide a clear image such as concentration to tumors in a mouse, which is a similar imaging ability to human PET. Thus, ETCC (CC) has a good imaging power over the wide energy dynamic range (200 - 1500keV) with a wide field of view (FOV, 3str), and then it is ensured that this camera has a potential of developing the new reagents for molecular imaging. Already we have carried out several comparisons of our images with those of SPECT and PET. Also, multi-tracer image using I-131 and FDG, and imaging test using several RIs (ex. ^{54}Mn ,^{65}Zn) emitting higher energy gamma rays than 511keV for small animals have been carried out successfully. Recently several new biomarkers including nano-particles for drug delivering approach were examined to verify the merits of ETCC for medical imaging. In addition, we will present the unique ability of ETCC to reconstruct 3D image only from one directional observation.

K206: Research and developments of the multiple-molecular imaging GREI and new molecular imaging probes

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Multiple molecular imaging, which allows simultaneous visualization of the molecular dynamics in various biological processes, is required to achieve more advanced and precise diagnosis. To realize the multiple molecular imaging, we have demonstrated the feasibility of semiconductor Compton cameras for multiple molecular imaging in nuclear medicine. The Compton camera used in this work comprises two double-sided orthogonal-strip germanium detectors, which we call “GREI (Gamma-Ray Emission Imaging)”. Their excellent energy resolution enabled discrimination of the nuclides and accurate determination of the scattering angle for γ rays in wide energy range. In parallel with the system development, it is also required that exploration of imaging probes which have suitable characteristics for the GREI. The throughput speed and sensitivity of the present prototype is not so powerful that approximately 10 hours of data acquisition is needed for a reasonable image reconstruction. To satisfy such requirement, we investigated an experimental model using tumor-bearing mice and antibody probes. For example, cetuximab, anti epidermal growth factor receptor (EGFR) antibody, was labeled with ^{64}Cu and injected to the EGFR-over expressing tumor-bearing mouse. The similar accumulation of ^{64}Cu in the tumor region was detected by PET and GREI. Using the GREI, the *in vivo* distributions of the multiple molecules are able to be visualized simultaneously by labeling with different radioactive metal nuclide.

K207: The production for nuclear medicine at the Research Center for Nuclear Physics Osaka University

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Recently, positron emission tomography(PET) has been remarkably developed for visualizing the distribution of a radionuclide the human and animal’s body. Now the system is widely used for the diagnosis of cancer. Clinical application of PET has been made possible with the development of ingenious techniques for rapid synthesis of radiopharmaceuticals, suitable for in vivo studies, using cyclotron within a medical complex.

We selected to produce the radioisotope-labeled hematoporphyrin for the PET although the radio-pharmaceutical mostly used is the ^{18}F-labeled fluoro-deoxy-glucose which is similar to glucose in behavior, We synthesized ^{52}Fe($T_{1/2}$=8.28h), ^{62}Zn($T_{1/2}$=9.3h) and ^{124}I($T_{1/2}$=4.18d) which were positron emitters by the ^{52}Cr(^{3}He,3n)^{52}Fe, ^{62}Cu(p,n)^{62}Zn and ^{124}Te(p,n)^{124}I reaction, respectively with the AVF cyclotron at Research Center for Nuclear Physics, Osaka University.

The PET imaging used ^{62}Zn and ^{124}I were obtained and further we plan to them using porphyrin compounds.

K208: RI production in RIKEN for medical and pharmacological studies.

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In RIKEN Nishina Center for

Accelerator-Based Science, we have established production methods of various radioisotopes (RIs) including ^{48}V, ^{65}Zn, ^{89}Zr, and ^{109}Cd by use of the RIKEN Ring Cyclotron and AVF Cyclotron. These RIs have been used for tracer studies in medicine, pharmacology, biology, nuclear chemistry, and so on. On the other hand, we have developed a semiconductor Compton camera called "GREI", which enables the simultaneous visualizing of RI distributions in wide gamma-ray energy range. In addition, because of a number of biomolecular medicines have developed and are considered as molecular imaging probes, there is growing interest for production of intermediate-half-life RIs. In RIKEN Center for Molecular Imaging Science, we have established the ^{64}Cu (T1/2=12.7 hour) production method using a small cyclotron. Moreover, we have started to develop the ^{65}Zn and ^{89}Zr production method. The outlines of these RI production methods and imaging applications in RIKEN will be discussed.

K209: Global Medical Isotope Crisis threatens Nuclear Medicine Diagnoses (^{99m}Tc)

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Two of the most important isotope producing reactors, NRU reactor in Canada and HFR reactor in the Netherlands, are currently shut down. The production of more than 95% of the available Mo-99 has been performed in only five reactors in the world including above two reactors. Tc-99m is used in about 65% of all diagnostic nuclear imaging procedures, corresponding to about 0.9 million procedures yearly in Japan. NRU and HFR are expected to come back online at latest in coming September. The current medical isotope crisis will be alleviated after the two reactors returning to service. However, all of these five reactors are approximately 50 years old. Because the remaining life time of these reactors is not long, new reactors being built or new technology being developed, such as accelerator methods or the neutron capture of Mo-98 in a reactor, are proposed for the security of supply of medical isotopes.

In addition to a problem of the production rate, the methods using the molybdenum target lead to much lower specific activity Mo-99 compared to the fission technology. This would not work with current Tc-99m generators and would cause a problem with Tc-99m imaging kits. The new chemical processing technologies, manufacturing Tc-99m radiopharmaceuticals from such low specific activity Mo-99, are more importantly expected.

K210: Radiochemistry and medical radioisotopes produced by accelerator neutrons

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^{99m}Tc with a half-life ($T_{1/2}$) of 6 h, the daughter nuclide of ^{99}Mo with $T_{1/2}$ = 66 h, has been most widely used in nuclear medicine for diagnostics. So far, various reactions such as ^{100}Mo(p,pn)^{99}Mo, ^{100}Mo(p,2n)^{99m}Tc, and ^{100}Mo(γ,n)^{99}Mo were studied to produce ^{99}Mo or ^{99m}Tc. Recently, we proposed a new route to produce ^{99}Mo via ^{100}Mo(n,2n)^{99}Mo using fast neutrons from an accelerator. Note that the (n,2n) reaction cross section is quite large, ~ 1.5 b at neutron energy E_n ~ 14 MeV, cross sections for radioactive waste production reactions, such as (n,α), (n,np), (n,p), and (n,γ), at E_n ~ 14 MeV are quite small, and intense ~ 14 MeV neutrons could be obtained using a small accelerator. In addition, a significant progress has been achieved in accelerator technology, which enables us to obtain high flux fast neutrons. These features could allow us to obtain a significant amount of ^{99}Mo using the proposed method. We could also obtain various medical radioisotopes using fast neutrons from accelerators. I will discuss our works of medical isotope production and an important role of radiochemistry in the medical application of the isotopes.

1A01: Nuclear deformation dependence of the excitation functions of heavy ion fusion reaction in the ^{139}La+^{16}O, ^{159}Tb+^{16}O and ^{169}Tm+^{16}O systems

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We have performed experiments for the syntheses of heavy elements in the ^{nat}La + ^{16}O, ^{169}Tm + ^{16}O and ^{159}Tb + ^{16}O systems to discuss the effect of nuclear deformation of targets having different deformations. The evaporation residue cross sections were measured by using a stack technique and / or a gas-jet technique. The cross sections were derived from radioactivities of the products and used for constructing excitation functions of nuclear fusion. It was found that the threshold in the fusion reaction gets lower for the deformed targets. This may be because the coulomb barrier changes according to configuration of the deformed targets and the projectile. Further discussion will be made with the theoretical calculation with the ALICE code and the HIVAP code.

1A02: Fission fragment anisotropy in heavy-ion-induced fission of actinides

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In heavy-ion-induced fission of actinides, fission fragment anisotropy has been found to become anomalously high compared with that expected from the standard saddle model. The pre-equilibrium *K*-state model suggests that entrance channel mass asymmetry $\alpha=(A_t-A_p)/(A_t+A_p)$ plays an important role for fission fragment anisotropy in heavy-ion-induced fission of actinides. However, a limited number of experiments have been carried out to study the effects of different entrance channels leading to the same compound nucleus. In order to study the effects of entrance channel mass asymmetry α on fission fragment anisotropy, we measured fission fragment angular distributions in the reactions of ^{22}Ne+^{232}Th (α=0.827) and ^{12}C+^{242}Pu (α=0.906), populating the same compound nucleus ^{254}Fm. At the JAEA tandem accelerator fission fragments from the compound nucleus at excitation energies of E_{ex}=56.1, 52.5, 48.8 and 45.2 MeV were measured by four position-sensitive parallel-plate avalanche counters and a silicon surface barrier detector. The measured center-of-mass angular distributions of fission fragments $W(\theta)$ provides the fission fragment anisotropy defined as $A=W(180^o)/W(90^o)$. The anisotropy for ^{22}Ne+^{232}Th with small entrance channel mass asymmetry α=0.827 shows large values compared with those for ^{12}C+^{242}Pu with α=0.906. This implies that entrance channel mass asymmetry plays a role for fission fragment anisotropy in the present systems.

1A03: Cross-section measurement for neutron-induced fission of minor actinides using Kyoto University lead slowing-down spectrometer

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The neutron-induced fission cross-sections for minor actinides (MA) have been measured in the neutron energy range from 0.03eV to 2keV using the lead slowing-down spectrometer (KULS) at the Research Reactor Institute, Kyoto University. The KULS is a lead cube of 1.5x1.5x1.5m^3. Neutrons are produced by irradiating tantalum plates located in the center of the KULS with a 30-MeV pulsed electron beam. The neutrons are slowed down by inelastic and elastic scatterings and finally reach to a MA sample. The neutron energy was determined from the time-of-flight in the lead material. Although the energy resolution is about 40%, a high neutron flux is available using the KULS since the sample can be irradiated at a small distance, typically 40cm, from the neutron production target. Up to now, the cross-sections for ^{237}Np, $^{241,\ 242m,\ 243}$Am, $^{245,\ 248}$Cm were obtained. In the presentation, the results of the measurement, mainly for ^{248}Cm will be shown.

Present study is the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

1A04: Production and decay properties of ^{261a,b}Rf

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Relatively long-lived isotope of Rf, ^{261a}Rf ($T_{1/2}$ = 68 s), has been used in chemistry experiments of

Rf. In recent years, it is said that a short-lived isomer of ^{261}Rf, ^{261b}Rf ($T_{1/2}$ = 3 s), exists. Although this spontaneously fissioning 3-s activity was known previously, it had been assigned to ^{262}Rf. The aim of the present work is to clarify existence of the short-lived isomer of ^{261}Rf by direct production of Rf isotopes.

Experiments were performed at the RIKEN linear accelerator (RILAC) facility using a gas-filled recoil ion separator, GARIS. Rf isotopes were produced in the reaction of ^{18}O on ^{248}Cm. A $^{248}\mathrm{Cm_2O_3}$ rotating target having a thickness of 230 μg cm^{-2} was irradiated by ^{18}O beams of energies of 88.2, 90.2, 94.9, and 101.3 MeV at the middle of the target. Produced nuclides in the ^{248}Cm(^{18}O, xn)$^{266-x}$Rf reaction were separated from the beam by GARIS, and implanted into a position sensitive detector which mounted on the focal plane of GARIS. The beam was switched ON–OFF repeatedly at constant time intervals, and the measurements were performed only in the beam OFF period. In order to distinguish the decays of ^{261b}Rf and ^{262}Rf ($T_{1/2}$ = 47 ms), beam ON–OFF time intervals were set to 6 s–6 s and 0.1 s–0.1 s, respectively.

1A05: Production of ^{265}Sg via the ^{248}Cm(^{22}Ne,5n)^{265}Sg reaction

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We are developing a gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator GARIS to start superheavy element chemistry in RIKEN. In this work, we have investigated the performance of the system using an isotope of element 106, ^{265}Sg, produced in the ^{248}Cm(^{22}Ne,5n) reaction. Alpha-particles of ^{265}Sg separated with GARIS and extracted to a chemistry laboratory were successfully identified with a rotating wheel system for α-spectrometry under low background conditions. Based on time-correlated 24 α-α(-α) and 18 α-SF (Spontaneous Fission) events, two isomeric states in ^{265}Sg, ^{265a}Sg (α energy E_α = 8.84 MeV, half-life $T_{1/2}$ = 6.7 s) and ^{265b}Sg (E_α = 8.69 MeV, $T_{1/2}$ = 15 s), were identified. The decay patterns of ^{265}Sg in the chain ^{265}Sg → ^{261}Rf → (^{257}No →) were revised together with those of ^{261}Rf in our previous ^{248}Cm(^{18}O,5n) experiment. The cross sections for the ^{248}Cm(^{22}Ne,5n)^{265}Sg reactions at 118 MeV were roughly evaluated to be 200 pb for ^{265a}Sg and 170 pb for ^{265b}Sg. In the symposium, the results of the production of ^{90}Mo and ^{173}W via the ^{nat}Ge(^{22}Ne,xn)^{90}Mo and ^{nat}Gd(^{22}Ne,xn)^{173}W reactions, respectively, will be also presented for future chemistry experiments of Sg.

1A06: α-HiB complexation and ion-exchange behavior of the group-4 elements as homologues of Rf.

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α-HiB complexation and cation-exchange behavior of Zr, Hf, and Th as homologues of Rf have been studied by a batch method using the carrier-free radiotracers ^{88}Zr, ^{175}Hf and ^{234}Th. It was found that the cation-exchange behavior of Zr, Hf, and Th in α-HiB / $\mathrm{HNO_3}$ reaches equilibrium at 180 min under the given conditions. The distribution coefficients (K_d values) of Zr and Hf on the cation-exchange resin at [H$^+$] = 0.1 M decreased with an increase of [α-HiB$^-$], reflecting successive formation of α-HiB complexes of these elements. In contrast, Th and Y which is used as a representative of the trivalent ion were strongly adsorbed on the resin under the same conditions, obviously showing different behavior from that of Zr and Hf.

1A07: Sulfate complexation of element 104, Rf, in $\mathrm{H_2SO_4/HNO_3}$ mixed solution ([H$^+$] = 1.0 M)

LI, Z. J.[1], TOYOSHIMA, A.[1], ASAI, M.[1], TSUKADA, K.[1], SATO, T. K.[1], SATO, N.[1], KIKUCHI, T.[1], SCHÄDEL, M.[1], NAGAME, Y.[1], LIANG, X. H.[2], KASAMATSU, Y.[3], KOMORI, Y.[3], OOE, K.[3], SHINOHARA, A.[3], GOTO, S.[4], MURAYAMA, H.[4], MURAKAMI, M.[4], KUDO, H.[4], HABA, H.[5], TAKEDA, Y.[6], NISHIKAWA, M.[6], YOKOYAMA, A.[6], IKARASHI, S.[7], SUEKI, K.[7], AND AKIYAMA, K.[8]

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Cation-exchange behavior of ^{261}Rf ($T_{1/2}$ = 78 s) produced in the ^{248}Cm(^{18}O, 5n) reaction was studied on a “one-atom-at-a-time” scale in H_2SO_4 (0.15−0.69 M)/HNO_3 mixed solutions ($[H^+]$ = 1.0 M) using an automated ion-exchange separation apparatus coupled with the detection system for alpha-spectroscopy. It was found that adsorption probability (%ads) of ^{261}Rf on cation-exchange resin decreases with increasing $[HSO_4^-]$, showing a successive formation of its sulfate complexes. Rf exhibited much weaker formation of the complexes than the lighter homologues Zr and Hf, which is qualitatively in good agreement with theoretical calculations including relativistic effects.

1A08: On-line isothermal chromatographic behavior of the group-5 elements Nb and Ta as homologues of Db

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To investigate chemical properties of the transactinide element dubnium (Db, Z = 105), we have so far developed an on-line isothermal gas chromatographic apparatus. In this work, the on-line experiments with group-5 elements, Nb and Ta, using short-lived isotopes ^{88}Nb and ^{170}Ta as the homologues of Db were carried out. We determined optimum conditions to produce volatile compounds of the elements. Under the obtained conditions, overall efficiencies of the volatile compounds were measured as a function of the isothermal temperature in chlorinating condition with mixture of nitrogen gas and the $SOCl_2$ vapor containing 1% oxygen. Maximum yields of Nb and Ta were about 10 % and 15 %, respectively. Based on the isothermal gas chromatographic behavior of them, it was found that Ta compounds would be less volatile than Nb ones.

1A09: Research on gas-phase chemistry for chloride of group-4 elements

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A gas-phase chemical separation is one of the most utilized techniques to study chemical properties of superheavy elements (SHE). SHE’s produced in nuclear reaction are converted to volatile compounds by chemical reaction. The volatility of SHE compounds is determined by their adsorption-desorption processes on an isothermal chromatographic column. In the present work, we investigated gas chromatographic behavior of volatile chloride compounds of group-4 elements using a fabricated apparatus for the study of Rf.

The isotopes of Zr and Hf were produced in the ^{nat}Ge(^{18}O,xn) and ^{nat}Gd(^{18}O,xn) reaction, respectively, at the JAEA tandem accelerator facility. Nuclear reaction products attached on a carbon cluster were transported to the gas chromatographic apparatus by a He carrier gas flow.

The transported products were collected on quartz wool plugged in a quartz tube where a reactive HCl gas was added to form volatile chloride compounds. The formed volatile compounds were then fed into an isothermal chromatographic quartz column which is directly connected to the quartz tube. The volatile compounds of Zr and Hf passing through the isothermal column were collected on a glass filter at a collection site. The efficiency of the transportation of nuclear reaction products and the formation of volatile compounds were determined by γ-ray spectrometry using a HPGe detector.

The transportation efficiency was about 50%, and the combined efficiency of formation and re-transportation was about 20%.

Finally, the isothermal chromatogram of Zr and Hf volatile compounds were measured precisely.

1A10: Electrochemical oxidation of nobelium using a microchannel-electrode chip

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An electrochemical oxidation experiment of nobelium (No), element 102, has been carried out using a microchannel-electrode chip which was a microchip integrated with electrodes. The isotope ^{255}No ($T_{1/2}$ = 3.1 min) was produced in the ^{248}Cm(^{12}C,5n) reaction using the RIKEN K70 AVF cyclotron. The reaction products recoiling out of the target were transported by a He/KCl gas-jet transport system to a chemistry laboratory. The transported products were then dissolved in 1 μL of 0.1 M HNO_3. The solution was fed into the microchannel-electrode chip and the potential of 500 or 1250 mV vs. Ag pseudo-reference electrode was applied to the working electrode. The effluent was mixed with 0.5 M di(2-ethylhexyl) phosphoric acid in CCl_4 solution and the mixture was shaken for 1 min to separate No^{3+} from No^{2+}. The activities of ^{255}No in both aqueous and organic phases were determined by α-spectrometry using Si detectors. The oxidation of No^{2+} to No^{3+} was observed at the applied potential of 1250 mV, while at the applied potential of 500 mV, No^{2+} was not oxidized.

1A11: Relationship between particle size distributions of radioactive aerosols and their half-lives in an accelerator facility

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In accelerator tunnels, aerosols formed by radiation were observed in the air. The radiation causes chemical and nuclear reactions, and the radiation-induced species are considered to be the source of the aerosol particles. The aerosols in the facilities often incorporate various radionuclides. The decay of the nuclides might influence the particle size distribution. The Monte Carlo simulation of the distribution was carried out to elucidate the relationship between the half-lives of the radionuclides and the particle size distribution. It was assumed that initial particles had a lognormal size distribution and that some of the aerosol particles incorporate the radioactive nuclide. The first simulation only took account of growth by condensation. The second simulation took account of the attachment of radioactive nuclide to the aerosol particle in addition to the first condensation model. The growth of the aerosol particles and the decay of the radionuclides in the aerosol were calculated. The particle size distribution based on the activity was also evaluated.

1A12: Temperature dependence of In-beam Mössbauer spectra of ^{57}Mn implanted into magnesium oxide

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A study of ^{57}Fe in-beam Mössbauer spectroscopy using ^{57}Mn provides direct information on exotic state of unstable nuclear probes and on electronic properties of the surrounding materials at the atomic scale. Recently, our detection method was successfully improved to obtain in-beam Mössbauer spectra with very large S/N ratio using HIMAC (Heavy Ion Medical Accelerator in Chiba) by developments of the PPAC detector and the anti-coincidence detection method. Here, we have measured the emission Mössbauer spectra of ^{57}Mn implanted into a simple metal oxide solid MgO of which temperature was controlled. The ^{57}Mn ions were produced as the projectile fragments of the high-energy ^{58}Fe beam. The sample was an MgO polycrystalline disk (50 mm diameter and 5 mm thickness) purchased from Furuuchi Chemical Co. The emission Mössbauer spectra of ^{57}Mn implanted into the MgO disk were obtained at the temperature range from 61 K to 623 K. Based on the density functional calculations, a singlet peak (I.S. = -0.94 mm/s) is assigned to high-spin Fe^{2+} located at a substitutional site of Mg^{2+} in MgO crystal. The other doublet components are considered to be Fe atoms located the Mg substitutional sites with vacancies of Mg or O atoms. Details of temperature dependence of obtained Mössbauer spectra will be discussed.

1A13: Temperature dependence of Mössbauer spectrum of ^{57}Mn implanted into sodium fluoride

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^{57}Mn in-beam Mössbauer spectra have been measured for sodium fluoride at 293K, 373K, 473K, 623K and 773K in order to elucidate the chemical and physical behaviors of trace transition metal impurity in a simple ionic alkali halide of rock salt type crystal structure. A singlet Mössbauer peak was observed at all temperatures for the substitutional ^{57}Fe site at the Na$^+$ site of NaF. Two additional doublet species were observed and interpreted as ^{57}Fe site with one or more crystal defects around the ^{57}Fe. At higher temperatures the population of the defect sites decreased and the singlet component increased, suggesting that the defects were annealed and disappeared.

1A14: Studies of Giant Hyperfine Magnetic Fields in Linear Two-Coordinate Fe Complexes

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There has been interested in transition-metal complexes with lowering symmetry of unsaturated linear two-coordination. It is reported that some of these complexes exhibit unique behavior in physical and chemical properties, notably unusual magnetism. Recently, we succeeded in synthesizing a linear coordinate Fe complex with a newly developed "Rind" ligands, Fe[MPhindS]$_2$ and Fe[MPhindNH]$_2$. "Rind" (R=Me, Ph) is based on a fused-ring *s*-hydrindacenyl structure [1,2]. The X-ray structure analysis, magnetization measurement, ^{57}Fe Mössbauer spectroscopy, and μSR were performed to elucidate the physicochemical property of Fe[MPhindS]$_2$ (S–Fe–S=161.4º) and Fe[MPhindNH]$_2$ (N–Fe–N=179.1º).

Mössbauer absorption spectra were measured by a conventional method between 1.6 K and RT under applied zero-field. The obtained Mössbauer spectra could be analyzed a single component of symmetric doublet down to 1.6 K. The derived values of *I.S.* and *Q.S.* at 1.6 K were 0.83(1) mm/s and 1.04(2) mm/s, respectively. It was assigned from these Mössbauer parameters that Fe atoms of Fe[MPhindS]$_2$ are in high-spin 2+ state. However, the linewidth (FWHM) was not strongly dependent on temperature, and there was no evidence of hyperfine magnetic splitting or relaxation pattern at the lowest temperature. On the other hand, the Mössbauer spectra of Fe[MPhindNH]$_2$ revealed the magnetic relaxation below 77 K. We will discuss the origin of the giant hyperfine magnetic fields of linear two-coordinate Fe complexes.

Ref.:
[1] A. Fukazawa *et al.*, *J. Am. Chem. Soc.*, **129**, 14165 (2007).
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1A15: Iron nitride films with high nitrogen content produced by laser deposition

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Iron nitride has been studied extensively, because it can exist in many different phases with different structures and properties with various nitrogen contents. Paramagnetic phase with a stoichiometry of FeN were recently discovered; γ’’-FeN and γ’’’-FeN. But only a mixture was found, and the single phases are yet to be obtained. We performed PLD of Fe metal in N_2 atmosphere to produce iron nitride films deposited onto Al substrates. The synthesized films were investigated by Mössbauer spectroscopy, powder XRD and SEM.

Iron nitride films were produced onto Al substrates at room temperature varying N_2 pressure; changing the nitrogen content of the films. When the nitrogen pressure was kept at 70 Pa, γ’’-FeN (δ = 0.07 mm/s) and γ ’’’-FeN (δ = 0.63 mm/s) were produced. The substrate temperatures while PLD was changed, while the nitrogen pressure was kept at 70 Pa. The sample produced at 100 K had γ’’-FeN as dominant products, and the yield of γ’’’-FeN enhanced by increasing the substrate temperature at 300 K. At the highest temperature at 520 K, γ’’’-FeN became dominant and γ-Fe appeared. In this study, it was demonstrated that the yields of γ’’-FeN and γ’’’-FeN were controlled by changing the substrate temperature in PLD.

1A16: Dilute magnetism and Mössbauer spectra of nano SnO$_2$ powders co-doped with Fe and V ions

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([1]Tokyo University of Science., [2]The University of Tokyo.)

The magnetic properties of SnO_2 are enhanced with doping Fe concentration, and on the contrary, the magnetic moments decrease with doping V concentrations. In order to clarify the dilute magnetism and to control the magnetic properties, we study on co-doping effect of SnO2 with V and Fe ions by Vibration Sample Magnetometer and Mössbauer spectroscopy. The mixed powders of SnO_2:Fe_nV_X (n=1, 2, x=0.5 ,1 ,2) were prepared by a sol-gel method using citric acid and ethylene glycol. The samples were annealed finally at 550°C for 2 hours. XRD patterns, which were not so affected by co-doping, showed only Rutile structure of SnO_2. SnO_2 doped with 0.5%V and 1%Fe showed the strongest ferromagnetism in those series. In the Mossbauer spectrum, the magnetic sextet with B_{hf}= 50.7 T, δ= 0.38 mm/s, and $2\varepsilon = - 0.29$ mm/s and Δ =1.14mm/s was observed in addition to two Fe^{3+} doublets, whereas Mössbauer spectra of the other samples consisted of magnetic relaxation peaks and paramagnetic doublets. The bulk magnetic properties may be correlated with the magnetic sextet.

1B01: Measurements of Mercury and Gold concentrations in samples from tailing dams which contaminated by Small Scale Mining, using PGAA and INAA

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Small scale gold mining is widely seen in many places in the world. During gold extract processing, large amount of mercury was released in the environment. Small-scale gold miners in the Philippines use orders of magnitude more mercury for extracting gold than small-scale miners in most other countries. In the order of 145 tons of mercury is released per year from some of the small-scale mining communities. Extrapolated to the whole country 200 to 500 tons of mercury is released to the environment every year. Most of the mercury will end in the mangrove swamps fringing the coast line. The mangrove swamps serve as hatching grounds for fish and shellfish the main source of proteins for the Filipinos. Very high concentrations of mercury have been found in water which was used for watering rice paddies.

In this study, we developed determination of mercury and gold in the crushed rock mines nondestructively using PGAA and INAA. Also based on the concentration of gold and mercury in tailings, we will optimize conditions of "the state battery method". Observed mercury and gold concentrations in tailings are 2.5 – 393 ppm, 1-3.9 ppm, respectively. Based on these results, we will optimize the conditions of the state battery method for mercury clean up project.

1B02: Estimation of anthropogenic influence on atmospheric environment by analyzing particulate matter in snow cover

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Atmospheric aerosols, derived from whether natural or anthropogenic origin, work as ice nuclei in atmosphere and are captured in snow. They are conserved in snow cover, which shows the state of atmospheric environment at the time of snowfall. Therefore, analyzing particulate matter in snow cover, we can evaluate average atmospheric environment of one winter season, not that of the moment of sampling.

We collected snow samples at Hokuriku Region, Kanto Region and Hokkaido from 2008 to 2010, considering the factor of snowfall, effect of northwest monsoon and existence of local contaminant. The snow samples were melted at room temperature and filtered with φ0.22μm membrane filter to separate particulate matter in snow. Because particulate matter in snow cover has a characteristic of low concentration and high elemental diversity, we applied multiple prompt gamma-ray analysis (MPGA) in addition to instrumental neutron activation analysis (INAA) to elemental analysis. The net weight of particulate matter on the membrane filter is hard to measure, so we calculated enrichment factor (EF), instead of concentration of respective elements, to discuss the result.

1B03: Bulk chemical analysis of hydrogenous

meteorites by neutron induced prompt gamma-ray analysis

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Meteorites are generally heterogeneous in chemical composition. Bulk chemical analysis of meteorites offers a challenge for representative sampling. In practice, increasing sample mass introduce neutron flux perturbation and gamma-ray self attenuation inside the sample. Moreover, the analytical sensitivity of the elements changes in the presence of strong scatterer like H in the sample. Among the recovered meteorites CI and CM carbonaceous chondrites contain the high abundance of H (up to 2 wt %). In this study, we aimed to determine element contents in the carbonaceous meteorites by using neutron induced prompt gamma-ray analysis (PGA). To evaluate analytical sensitivity variation of the elements with sample mass and H content, cold-pressed disk (12 mm diameter) samples were prepared using homogeneous mixture of GSJ basaltic rock JB-2 and oxalic acid with 2 wt % H. All samples and standards were irradiated by guided beam of thermal neutrons at JRR-3M of JAEA. The analytical sensitivity of the elements remains constant within the error of counting statistics when a disk mass increases up to 0.5 gm and after that decreases with increasing of the disk mass. Therefore, in PGA analysis of hydrogenous meteorites (~2 wt% H) up to 0.5 gm sample, the uncertainty due to the analytical sensitivity changes can be ignored.

1B04: Simple and high-precision measurement method of neutron distribution using ink-jet printer

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Neutron flux is an important parameter in neutron activation analysis, radioisotope production and other neutron irradiation experiments. In the case of neutron activation analysis, the analytical accuracy depends on the estimation of neutron flux at irradiations. In order to measure neutron flux accurately, simple and high-precision measurement method of neutron distribution using an ink-jet printer has been developed. In this method, neutron monitors are prepared by applying gold solution on a paper sheet using an ink-jet printer. The prepared monitor is thin enough to ignore the self-absorption in neutron irradiation and self-shielding in gamma-ray measurement, and is also easy to handle. Measurements of axial neutron distribution at the positions of irradiating capsules in neutron irradiation holes of HANARO, PTS#1 and PTS#3, were tried using the neutron monitors. As a result, a trend or fluctuation of neutron fluence cannot be observed in the case of PTS#1, but there is a positive trend in PTS#3. This result is consistent with the previous report of neutron flux measurements using Au-Al wire monitors. The result of measurements of neutron distribution using an imaging plate at neutron irradiation facility of KUR will be also presented.

1B05: A study on speciation of chromium in soil by X-ray absorption fine structure.

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Hexavalent chromium (Cr(VI)) is toxic, and one of the hazardous materials. 1,5-diphenylcarbazide spectrophotometry is widely used as determination of Cr(VI) contents in environmental samples. However, some problems are pointed out in this method, namely, there is a difficulty when applying to soil samples. In this study, we applied X-ray absorption fine structure (XAFS) spectroscopy to determination of Cr(VI) contents, that was one of useful means for nondestructive analysis. In the conventional methods, the normalization energy of the Cr K-edge in X-ray absorption near-edge structure (XANES) spectra was not clearly defined. So, it was difficult to determine Cr(VI) contents in interstitial water or soil samples with sufficient accuracy. Therefore, we investigated to find the best normalization energy in the XANES spectra.

It was considered that the peak near the white line in the spectrum of the standard reference was due to d-d transition of Cr (III). The peak observed near 6010 eV in white line is peculiar to Cr(III), ascribing to the transition of $^4A_{2g}\rightarrow{}^4T_{2g}$. By normalization at this position, we were able to obtain the better calibration curve (R^2=0.988).

1P01: Gamma-ray analysis and behavior of spallation products produced in mercury of the neutron source at J-PARC

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Neutrons are produced by spallation reactions by proton beam (3GeV, 1MW) irradiation on mercury in the Material and life science experimental facility at J-PARC. Mercury was designed to be drained into a shielded tank to reduce the radiation by the spallation products. However it was confirmed that some spallation products (mainly ^{188}Ir, daughter of ^{188}Pt) adhering to the inner surface increased the dose after the draining. To get more information the authors sampled mercury (~120 g) and the adhesive substances (black powder). Then, behaviors of the spallation products in mercury are discussed.

Twelve nuclides, ^{203}Hg, ^{188}Pt, ^{189}Ir, ^{188}Ir, ^{185}Os, ^{175}Hf, ^{173}Lu, ^{172}Lu, ^{146}Eu, ^{113}Sn, ^{110m}Ag, ^{88}Y, were identified in the mercury specimen by gamma-ray spectroscopy. The amounts of radioactivity were in the range between several to around hundred percents relative to calculation. Only ^{110m}Ag was not found in the adhesive substances. It is expected that other elements than mercury take important role to behavior of the spallation products since the content ratios of the elements were calculated to be in the order of ppb, and the elements (tin and europium) having higher solubility than silver were found in both specimens. For example, iron, which is expected to be a dominant impurity (caused by erosion of stainless steel pipes), dose not react with silver. On the other hand, iron reacts with tin.

1P02: Study on the reversed phase extractive chromatography in micro-columns for speciation of rutherfordium

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For the purpose of speciation of rutherfordium, a superheavy element, in solution, we aim to obtain liquid‐liquid extraction data of Rf by means of reversed‐phase chromatography with stationary phase containing chelate extractant, TTA. TTA extracts quadrivalent metal ions preferentially, and, that is, we can obtain a specific complex formation constant of Rf. In this work, we filled micro‐columns with stationary phase containing TTA, and measured percent adsorption of Zr and Hf, homologues of Rf, in the tracer scale as a function of [F$^-$].This results coincide with macro‐scale experiments in the following respects. Adsorption values decrease with increase of [F$^-$] and good separation between Hf and Zr is expected around [F$^-$]$_{eq}$=6.0×10^{-5} M. Therefore, the percent adsorption measurement with the micro‐columns is considered to be applied for the Rf experiment.

1P03: Study on the adsorption rate of Zr and Hf as homologues of element 104, Rutherfordium, on cation-exchange resin in H_2SO_4 solutions .

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([1]Pure & Appl. Sci., Univ. Tsukuba, [2]Advanced Sci. Res. Center., JAEA)

It was reported that the cation-exchange chromatographic behavior of Zr and Hf as homologues of element 104, Rf, in H_2SO_4 solutions was not consistent with the distribution coefficients obtained in the batch experiment. In the present study, the adsorption rates of Zr and Hf on the cation-exchange resin in H_2SO_4 solutions were investigated in order to lead to the quantitative discussion of the chromatography. It was obtained the adsorption rates of ^{88}Zr and ^{175}Hf on the cation-exchange resin (MCI GEL CK08Y, H^+ form) in 0.1 M H_2SO_4 at 15, 25 and 45℃ as functions of the shaking times. The results showed that the equilibrium of cation-exchange was reached at 70-80 seconds and that the adsorption rates on the cation-exchange resin were not changed between 15－45℃ in the present condition. In H_2SO_4/HNO_3 mixed solution ([H$^+$]$_{eq}$=1.0 M), the adsorption rate was similar to those of ^{88}Zr and ^{175}Hf on the cation-exchange resin in H_2SO_4 solution.

1P04: Development and evaluation of a liquid scintillation detector for aqueous chemistry of superheavy elements

KOMORI, Y.[1], OOE, K. [1], KURIYAMA, A. [1], TAKAYAMA, R. [1], KIKUTANI, Y. [1], KIKUNAGA, H. [2], KASAMATSU, Y. [1], YOSHIMURA, T. [1],

TAKAHASHI, N. [1], SHINOHARA, A. [1]
([1]Grad. School of Sci., Osaka Univ., [2]Nishina Center, RIKEN)

We are planning to develop an on-line liquid scintillation detection system coupled to an ion-exchange column or a liquid-liquid extraction apparatus for aqueous chemistry of superheavy elements. In this work, we have developed a liquid scintillation detector with α/β discrimination for on-line measurements and evaluated its performances. From the results, it has been found that the detector has a counting efficiency of approximately 96% for ^{241}Am and a FWHM of approximately 310 keV for the 4784 keV α-energy of ^{226}Ra at the sample volume of 1 mL. The energy resolution became worse with increasing the amount of the sample solution in the detector cell. Next, test of the on-line measurement with the developed detector was performed. Aqueous solution including ^{241}Am and ^{152}Eu was introduced into the detector cell together with an emulsifier scintillator and mixed by an electromagnetic stirrer under the detection chamber and stirring bar in the glass cell. After well mixing, α-particle detection with a good energy-resolution and a discrimination between α- and β-events have been achieved.

1P05: **Anion-exchange behavior of the group-6 elements in HF/HNO$_3$ mixed solutions - Towards the study of ion-exchange behavior of $_{106}$Sg –**

LIANG, X. [1,2], LI, Z., TSUKADA, K. [1], TOYOSHIMA, A. [1], ASAI, M. [1], SATO, T.K. [1], SATO, N. [1], KIKUCHI, T. [1,3], KANEYA, Y. [1,3], NAGAME, Y.[1]
([1]CIAE, China, [2]ASRC, JAEA, [3]Ibaraki Univ.)

Anion-exchange behavior of the group-6 elements, W and Mo, in HF and HNO$_3$ mixed solutions has been studied by a batch method using the carrier-free radiotracers ^{181}W ($T_{1/2}$ = 121.2 d) and ^{93m}Mo ($T_{1/2}$ = 6.9 h). The long-lived isotope ^{181}W was produced in the ^{181}Ta(p, n) reaction at the JAEA tandem accelerator. The ^{181}W isotope was chemically separated from the Ta target material, and was dissolved with 10^{-4} – 1.0 M HF and 0.1 M HNO$_3$ mixed solutions for batch experiments. On the other hand, the short-lived isotope ^{93m}Mo was produced in the ^{93}Nb(p, n) reaction. The recoiling products from the Nb target were transported to a chemical laboratory with a He/KF gas-jet system, and were dissolved with HF and HNO$_3$ mixed solution. The solutions containing ^{181}W or ^{93m} Mo were mixed with 5 – 200 mg anion exchange resin CA08Y. Radioactivities of the solution were assayed for γ-ray spectrometry, and then distribution coefficients (K_ds) of these elements were determined. It was found that the K_d values of W depend strongly on the concentration of HF, while those of Mo are almost constant as a function of [HF] under the given conditions.

1P06: Measurement of excitation function of ^{nat}Lu(*p,xn*)^{175}Hf

KIKUTANI, Y.[1], OOE, K.[1], KOMORI, Y.[1], FUJISAWA, H.[1], KURIYAMA, A.[1], TAKAYAMA, R.[1], KIKUNAGA, H.[2], KASAMATSU, Y.[1], YOSHIMURA, T.[1], TAKAHASHI, N.[1], SHINOHARA, A.[1]
([1]Grad. School of Sci., Osaka Univ., [2]Nishina Center, RIKEN)

We are planning to study the solvent extraction behavior of element 104, rutherfordium (Rf) and the lighter homologues of Rf, zirconium (Zr) and hafnium (Hf). The ^{88}Zr ($T_{1/2}$ = 83.9 d) and ^{175}Hf ($T_{1/2}$ = 70.0 d) nuclides are needed for the solvent extraction experiments of Zr and Hf. However, there is no data of the excitation function of ^{nat}Lu(*p,xn*)^{175}Hf reaction and the data is required for efficient production of ^{175}Hf. In this work, the excitation function of ^{nat}Lu(*p,xn*)^{175}Hf reaction at energies up to 14 MeV has been measured for the first time. Activation method and stacked foil technique were used to measure cross section of the ^{nat}Lu(*p,xn*)^{175}Hf reaction. The target stacks consisting of lutetium, iron and cupper foils were irradiated by a 14 MeV proton beam from the AVF cyclotron at Research Center for Nuclear Physics (RCNP), Osaka University. Gamma-rays of the radionuclides produced in these samples were measured with a germanium semiconductor detector. As a result, the obtained excitation function of the ^{nat}Lu(*p,xn*)^{175}Hf reaction shows the maximum value of 140 mb at the proton energy of about 10.3 MeV.

1P07: Measurement of Pa-231 photofission cross-section by the fission track method (II)

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Fission track method has been applied to the detection of photofission events of ^{231}Pa. For the detection of fission fragments, quartz and muscovite were used as solid state track detectors. The target nuclide ^{231}Pa was sandwiched between pieces of quartz and muscovite for irradiation with bremsstrahlug at a Linac facility of Tohoku Univ. together with the natural uranium (^{nat}U) and ^{232}Th targets used as references. After the irradiation, the detectors were subjected to chemical etching and observed for track counting using a microscope in order to evaluate the photofission cross-section of ^{231}Pa. At 20 MeV of the maximum bremsstrahlung energy, in our previous work, the fission probabilities of ^{231}Pa relative to ^{nat}U and ^{232}Th were determined to be around 2 and around 6, respectively, before. In the present study, the photofission probability of ^{231}Pa to that of ^{nat}U was determined to be around 2 at 18 MeV of the relevant energy. This result might exhibit a saturation behavior even down to the energy.

1P08: Development of a surface ionizer coupled to a gas-jet transport system for the measurement of the first ionization potential of Lr

SATO, N.[1], ASAI, M.[1], TSUKADA, K.[1], SATO, T.[1], TOYOSHIMA, A.[1], LI, Z. J.[1], KIKUCHI, T.[1,2], KANEYA, Y.[1,2], ICHIKAWA, S.[3], NAGAME, Y.[1], SCHÄDEL, M.[1]
([1]ASRC, JAEA, [2]Ibaraki Univ., [3]Nishina Center, RIKEN)

It is important to determine the first ionization potentials (IPs) of the heaviest elements with atomic number Z > 100 to understand their valence electronic structure affected by relativistic effects. Due to low production rates and short half-lives of the heaviest nuclides, IPs of these elements have not been so far determined. We are developing a surface-ionization-type ion source coupled to a gas-jet transport system in the Isotope Separator On-Line (ISOL) at the JAEA tandem accelerator facility to measure IP of the heaviest actinide, lawrencium (Lr, Z = 103). In the symposium, we report the present status of the development of the apparatus. Production yields of ^{256}Lr (half-life = 27 s) that is used for the IP measurement, in the ^{249,250,251}Cf(^{11}B, *xn*), ^{243}Am(^{18}O, 5*n*) and ^{248}Cm(^{14}N, 6*n*) reactions will be also presented.

1P09: Development of Si-CdTe detector array for Nuclear spectroscopy of superheavy nuclides

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Nuclear spectroscopy for understanding of nuclear structure is one of the hot topic in nuclear physic on the superheavy nuclides. Now, we are preparing the start-up of the nuclear spectroscopic study at the focal plane of GARIS or GARIS-II in RIKEN.

We pay attention to the CdTe detector as γ(X)-ray detector instead of the Ge detector.

The CdTe detector has strong points like as high absorption due to its high atomic number and high density, high energy resolution with low leak current due to wide band gap energy even at room temperature, and low cost to get the detector. Then, we developed a prototype of a new Si-CdTe detector array for alpha(e)-gamma spectroscopy of superheavy nuclides. Basic characteristics, such as energy resolution, time resolution, and background condition, of the Si-CdTe detector array was investigated by standard source of ^{241}Am and 3 mix source including ^{241}Am, ^{244}Cm, and ^{237}Np.

1P10: Gamma-ray analysis and behavior of spallation products produced in mercury of the neutron source at J-PARC

Kai, T., Kasugai, Y., Ooi, M., Wakui, T., Kogawa, H., Haga, K., Hanano, K.
(J-PARC center , Japan Atomic Energy Agency)

Neutrons are produced by spallation reactions by proton beam (3GeV, 1MW) irradiation on mercury in the Material and life science experimental facility at J-PARC. Mercury was designed to be drained into a shielded tank to reduce the radiation by the spallation products. However it was confirmed that some spallation products (mainly ^{188}Ir, daughter of ^{188}Pt) adhering to the inner surface increased the dose after the draining. To get more information the authors sampled mercury (~120 g) and the adhesive substances (black powder). Then, behaviors of the spallation products in mercury are discussed.

Twelve nuclides, ^{203}Hg, ^{188}Pt, ^{189}Ir, ^{188}Ir, ^{185}Os, ^{175}Hf, ^{173}Lu, ^{172}Lu, ^{146}Eu, ^{113}Sn, ^{110m}Ag, ^{88}Y, were identified in the mercury specimen by gamma-ray spectroscopy. The amounts of radioactivity were in the range between several to around hundred percents relative to calculation. Only ^{110m}Ag was not found in the adhesive substances. It is expected that other elements than mercury take important role to behavior of the spallation products since the content ratios of the elements were calculated to be in the order of ppb, and the elements (tin and europium) having higher solubility than silver were found in both specimens. For example, iron, which is expected to be a dominant impurity (caused by erosion of stainless steel pipes), dose not react with silver. On the other hand, iron reacts with tin.

1P11: GARIS-II commissioning #1

KAJI, D. [1], HABA, H. [1], KASAMATSU, Y. [1], KUDOU, Y. [1], MORIMOTO, K. [1], MORITA, K. [1], OZEKI, K. [1], SUMITA, T. [1], YONEDA, A. [1], SATO, N. [2], WAKABAYASHI, Y. [2], MURAKAMI, M. [3], TOKANAI, F. [4], NAMAI, S. [4]
([1]Nishina Center, RIKEN, [2]Advanced Science Research Center, JAEA, [3]Niigata Univ, [4]Yamagata Univ)

We developed a new gas-filled recoil ion separator GARIS-II for actinide-based asymmetric fusion (hot fusion) study. The 1st operation of GARIS-II was performed by using standard ^{241}Am source and 0 degree recoil atom of ^{208}Pb target produced by Rutherford scattering. The GARIS-II consists of five magnets arranged in a Q_v-D-Q_h-Q_v-D configuration. This configuration is the first design for the purpose of SHE study. At first, balance of magnet field strength was tuning for each magnet by standard ^{241}Am source with the activity of 21 kBq. A solid angle of GARIS-II was measured to be about 18.4 msr. This value almost reproduced an evaluated value by the computer code TRANSPORT. The solid angle is increased from 12.2 to 18.4 msr, approximately 1.5 times higher than GARIS.

The 2nd step of GARIS-II operation was investigated by 0 degree recoil atom of ^{208}Pb target produced by Rutherford scattering. Image size at focal plane of GARIS-II was investigated by changing the He gas-pressure in GARIS-II chamber.

1P12: Development of data acquisition system for short-time decay by using Flash ADC

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A data acquisition (DAQ) system has been upgraded for super heavy element research performed using the Gas-filled Recoil Ion Separator (GARIS).

Super heavy elements studied by using GARIS and GARIS-II, especially in the heaviest region on the nuclear chart, may have a very short life. The original GARIS readout electronics cannot separate sequential decays that occur at intervals shorter than 5 μs because of the pileup in the shaping amplifier. We modified the original readout system by using Flash ADC so that it can be used to detect shorter decay intervals.

The system was used in experiments to search for a new isotope ^{234}Bk in the reaction ^{197}Au(^{40}Ar,3n). A total of 119 decay chains were observed; all the decay chains were assigned to subsequent decays from ^{234}Bk. ^{218}Ra and ^{214}Rn, which have very short life times (15.6 μs and 0.27 μs, respectively) were contained in the decay chains. Sequential fast decay of these isotopes was clearly observed in the digitized preamplifier signal.

In this presentation, the modified DAQ system in detail as well as the observed data in the experiment search for the ^{234}Bk will be described.

1P13: Development of New Time of Flight Detector with a Large Diameter for Experiment of the Search for Superheavy Elements.

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A new time-of-flight (TOF) detector with a large effective area was developed for actinide-based asymmetric fusion (hot fusion) study by gas-filled recoil ion separators of GARIS and GARIS-II.

A system of time-of-flight (TOF) detectors and a position sensitive silicon detector (PSD) array are important roles for particle identification in fusion study. Especially, the TOF detectors are used for two purposes. One is to measure the time-of-flight of the particles coming into the PSD array to get the information of the mass number of recoil atom. The other is to distinguish decay signals in PSD array used in anti-coincidence mode.

In the case of hot fusion study, the image size in the focal plane of the separator becomes wide spread, because of multiple scattering of the recoil ion by the filling gas atoms. Therefore, we developed a new TOF detector with very large effective area of 120 mm in a diameter. As a first step of fundamental operation, a uniformity of detection efficacy was investigated by alpha particles from ^{241}Am standard source.

1P14: Development of an ionization chamber for nuclide identification of the superheavy elements

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([1]Tokyo Univ. of Science., [2]Nishina Center, RIKEN, [3]Osaka Univ.)

In the superheavy element region, the direct measurements of atomic number Z and mass number A of produced nucleus is one of the most challenging tasks. In the region of large Z, the density of electron-hole pairs is too high in using silicon detectors in the search experiments. As a result, it is thought that the precise measurement of the kinetic energy is not possible.

In this work, we operated ionization chamber with low pressure for reducing the density of ionization electrons and developed it for the purpose of precise measuring of the kinetic energy. We used alpha particles from an ^{241}Am source and performed performance evaluations so far. This time, we performed it with the ^{11}B beam at the tandem electrostatic accelerator in RIKEN.

We have measured the pulse height in each electrode while changing pressure. As a present result, measured values didn't accord with the values of the energy loss from calculation. It is thought that this because of the particle and the energy dependence of W-value. And the energy resolution was 2.13% in 4109.0 keV. We estimated standard deviations of mass for A=300 using this value and time resolution of Time-Of-Flight detector in the search experiments.

1P15: Study of Reactivity for Lanthanide Metallofullerenes Using Radiochemical Method

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Metallofullerenes are potential material for the construction of the drag deriver system which takes radioisotopes to the targeted organs. Our motivation is to synthesize the hydrophilic metallofullerene derivatives for the medical and pharmaceutical applications. In this session, we will report the synthesis and reactivity of metallofulleropyrrolidines for La, Ce, Pr, Nd, and Gd metallofullerenes.

Metallofullerenes samples were irradiated thermal neutrons in the JRR-3M research reactor of JAEA. These irradiated samples were refluxed in well dehydrated toluene with paraformaldehyde and N-methylglycine. After removal of residual reactant by distilled water, resulting solutions were condensed and injected into an HPLC column of Buckyprep. Eluate from the column was collected and detected their gamma-ray by Ge semiconductor detector.

Comparing HPLC chromatogram of "before reflux" with that of "after reflux", it was found that the intensity of the elution peak derived from La@C_{82} was decreased and that new peaks which derived from La@C_{82} derivatives were observed around 27 min and 55 min. Based on this result, HPLC elution peaks derived from metallofullerene derivatives of Ce, Pr, Nd, and Gd were also collected and detected their activity. As the results, we could not find significant differences in the reactivity between these metallofullerenes.

1P16: Separation of Metallofullerenes from hollow species using Lewis acid.

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Stevenson et al. reported that metallic nitride endohedral fullerenes and oxometallic fullerenes were separated from hollow species by using Lewis acids such as $AlCl_3$ or $FeCl_3$. In this study, we confirmed that this method could be applicable for the separation of metallofullerenes encapsulating lanthanide elements from hollow fullerenes. The extraction ratio of metallofullerenes was investigated as a function of weight of added AlCl3 and reaction time. Weight of $AlCl_3$ was varied from 5.1mg to 106.5mg with 30min reaction time. Reaction time was also varied between 5min to 3h with 15mg of $AlCl_3$. As a result, extraction ratio of metallofullerene was reached to be about 50% in the case of more than 25mg of $AlCl_3$ being added and of more than 30min of reaction time. The most of residual metallofullerene which could not be recovered is expected to be a insoluble substance such as polymerized metallofullerene. In conclusion, it is found that at least 50% of metallofullerenes were collected even if added excess quantity of $AlCl_3$ or took plenty of reaction time.

1P17: Recovery of molybdenum by using pyridine resin from simulant irradiated nuclear fuel solution

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There are many amounts of molybdenum in nuclear fuels irradiated in nuclear reactors. If molybdenum is immediately recovered after irradiation, we can obtain molybdenum-99. Molybdenum-99 is well-known as mother isotope of technetium-99m. Since the life time of molybdenum-99 is not so long, if we waiting several years after irradiation, the molybdenum without the radioactive isotope can be obtained. We have been studying the molybdenum recovery by using pyridine resin. First, the distribution coefficients of molybdenum and zirconium, which is known for accompanying elements with molybdenum, were evaluated in nitric acid solution and in hydrochloric acid solution. Second, the separation experiment of molybdenum and zirconium was investigated by chromatography. From result obtained by their investigation, we confirmed that the molybdenum can be recovered using hydrochloric acid system. Finally, we confirmed the recovery of molybdenum from simulant irradiated nuclear fuel solution by chromatography using pyridine resin in hydrochloric acid solution.

1P18: Synthesis and properties of hexatechnetium cluster complexes with mixed cap ligands of sulfide and chloride

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The octahedral hexanuclear cluster complexes having 24 valence electrons are known to show interesting photophysical and redox properties. In the recent study conducted by us, several chalcohalide-capped hexatechnetium(III) and hexarhenium(III) cluster complexes were synthesized and investigated about their chemical properties. In the present study, the new hexatechnetium(III) cluster complexes, $(PPh_4)_3[Tc_6(\mu_3\text{-}S_7Cl)Cl_6]$ and $(PPh_4)_3[Tc_6(\mu_3\text{-}S_6Cl_2)Cl_6]$ were synthesized and the physical properties were investigated. The complex was produced by the high temperature reaction of elemental ^{99}Tc, S, and $K_2[TcCl_6]$ with excess amount of KCl at 850℃. The reaction mixture was separated by column chromatography. Two different kinds of brown crystals, $(PPh_4)_2[Tc_6(\mu_3\text{-}S_6Cl_2)Cl_6]$ and $(PPh_4)_3[Tc_6(\mu_3\text{-}S_7Cl)Cl_6]$ were obtained. In acetonitrile solution, their UV-Vis spectra show the broad absorption bands of the cluster core with those of $(PPh_4)^+$. In addition, it was found that the spectrum of $(PPh_4)_3[Tc_6(\mu_3\text{-}S_7Cl)Cl_6]$ shifts to long-wavelength compared with that of $(Bu_4N)_3[Re_6(\mu_3\text{-}S_7Cl)Cl_6]$.

1P19: Production of no-carrier-added ^{177}Lu for radioimmunotherapy

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Lutetium-177 is considered to have potential for application in radioimmunotherapy, because it has favorable physical characteristics. We have succeeded in the production of high purity no-carrier-added (nca) ^{177}Lu of capable of labeling antibodies using reversed-phase ion-pair liquid chromatography. However, in this separation, nca ^{177}Lu could not be completely separated from more than 3 mg of Yb_2O_3 target. For this reason, the radioactivity of nca ^{177}Lu produced by our separation method is limited less than GBq. Therefore, for the realization of the large scale production, we considered the adoption of a coarse separation between Lu and Yb by using LnResin column before reversed-phase ion-pair liquid chromatography. In this study, the maximum weight of Yb_2O_3 capable of the separation between Lu and Yb by using LnResin column chromatography followed by reversed-phase ion-pair liquid chromatography was investigated. It was found that in less than 10 mg of Yb_2O_3 targets loaded on the LnResin column, the weight of Yb_2O_3 in the ^{177}Lu fraction was less than 2 mg. Therefore about 10 mg of Yb_2O_3 target was considered to be maximum weight of Yb_2O_3 capable of the separation between Lu and Yb by using LnResin column followed by reversed-phase ion-pair liquid chromatography.

1P20: Flux correction for photon Activation Analysis

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In photon activation analysis using bremsstrahlung from an electron LINAC, samples to be irradiated are stacked along an electron beam. Since photon flux comes down towards downstream samples, photon flux in each sample should be corrected to obtain an accurate determination value. We usually use Au foils for the flux correction, using ^{197}Au(γ, n)^{196}Au reaction. In this study we compared some metal foil monitors with Au monitor to get effectual correction way. Quartz filter samples with several kinds of monitors were irradiated by bremsstrahlung with 25 MeV of end point energy for 20 minutes. Decreasing rate of radioactivity in monitors was different from each induced radionuclides, and the rate was found to be correlated with Q value of a reaction producing radionuclide: the rate of decreasing became significant with decrease of absolute Q value. In the case of 420 minutes of irradiation time, the same systematic was also observed, but the difference of decreasing rate among monitors were smaller than the irradiation for 20 minutes. Appropriate monitor should be selected to obtain an accurate determination value.

1P21: Determination of trace amount of nickel by photon activation analysis

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Trace amount of nickel in high-purity metal samples was determined by Photon Activation analysis (PAA). It was confirmed that PAA was able to determine about 0.015 μg of nickel in the high-purity iron oxide sample within uncertainty of 20 % and that PAA used in this work made it possible to determine 1% of Ni at least in a few μg of magnetic spherules.

Photon activation analysis was carried out using the Electron Linear Accelerator at the Research Center for Electron Photon Science, Tohoku University. Samples, reference standards and gold foils as the fluence rate monitor of photons were enclosed in a quartz tube for irradiation with bremsstrahlung photons. The accelerator was operated at electron energy of 30 MeV with a mean current of around 0.11 mA for 8-10 hours. The quartz tube was placed just after a platinum converter and cooled with running tap water.

Then, applying PAA to the determination of trace amount of nickel in cosmic spherules, nickel was able to be determined by PAA more accurately than by instrumental neutron activation analysis which has so far been applied to analysis of spherule samples.

1P22: Hydrotreating of Kuwait Atmospheric Residue - Structural analysis of Nickel and Vanadium by XAFS Spectroscopy

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Nickel and vanadium are the most abundant trace metals in petroleum. Typically, metal-containing compounds are high boiling and entrained in the distillation bottoms (residuum) of crude. Additionally, metal complexes concentrate them in the asphaltene fraction of the residuum. To convert from residuum to clean fuels with higher value-added, it is necessary that included these metals remove from the residuum and apply residuum to Hydrodesulfurization (HDS). However, the metals (nickel and vanadium) that could not be removed are causes of disturbing the catalytic function used with HDS.

The objective of this study is that the local structures of Ni and V in Kuwait asphaltene, the following HDS catalyst are measured by XAFS spectroscopy. In the result, nickel and vanadium are suggested that similar with porphyrin-V and porphyrin-Ni in asphaltene, distinct from chemical state in catalyst.

1P23: Development of ^{41}Ca-AMS at University of Tsukuba

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Accelerator mass spectrometry (AMS) is an useful tool for low level measurement of long life radioisotopes. In the AMS, high terminal voltage is required for a sufficient isobaric separation with increase of atomic number. AMS of calcium-41 is not carried out in Japan and has a potential in dating and medical science. ^{41}Ca-AMS using the 12 UD peletron tandem accelerator at University of Tsukuba is appropriate measurement for the background. We have been developing ^{41}Ca-AMS system.

Commercial reagents of CaF_2 and CaH_2 were mixed with some materials used for a pilot beam which controls terminal voltage of the accelerator. From mixed sample of CaF_2 and ZnO, 14 nA of $^{40}Ca^{19}F_3^-$ and 23 nA of $^{66}Zn^{16}O_2^-$ (pilot beam of $^{40}Ca^{19}F_3^-$) were extracted. Besides, from sample of CaH_2 and MgO, 4 nA of $^{40}CaH^-$ and 2 nA of $^{26}Mg^{16}O^-$ (pilot beam of $^{40}CaH_2^-$) were extracted. On the other hand, from sample of CaH_2 which was irradiated using neutron and LiCl, 0.5 nA of $^{40}CaH_3^-$ was extracted, and 147 counts of ^{41}Ca were detected in 15 minutes using a ΔE-E counter. The ^{41}Ca/^{40}Ca atom ratio was estimated to be 3.1×10^{-8} from the beam current of $^{40}CaH_3^-$ and count rate of ^{41}Ca.

1P24: Determination of metallic elements in eye cosmetics by instrumental neutron activation analysis

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There are a lot of cheap cosmetics in the market, meanwhile these adverse effect on health is frequently overlooked. Colorful cosmetics particularly for eye makeup often have toxic heavy-metal elements as pigments. In this work, for the purpose of the determination of metallic elements in cosmetics and the estimation of toxicity of them, the eye cosmetics were analyzed by instrumental neutron activation analysis (INAA). We determined the concentrations of Mg, Al, Sc, Ti, V, Cr, Mn, Fe, Co, Zn, Cs and Ta in 30 samples of eye-shadow.

The concentrations of Al, Cs and Sc were constant within 30% in relative error. It suggests that these elements are included in major components. There were correlation between the concentrations of Fe, Mn and Co which were more rich in dark brown or black samples than white or whitish samples. All data of the concentration of Zn were able to be divided into two groups, below 50 ppm and several thousand ppm. Zinc values were different between packages made by respective producer. One sample in viridian color shows extremely high Cr concentration of about 1% which is 1000 times higher than that of the other samples. A chromium pigment might be included in this sample.

1P25: Improvements in precision of measurement of carbon isotope ratio with accelerator mass spectrometry

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In order to obtain a stable ion beam for measurement of precise isotope ratio of carbon by accelerator mass spectrometry, three improvements to JAEA-AMS-TONO were performed.

1. A cesium-beam sputter source is used to produce the ion beam of carbon. Cesium vapor is provided from a reservoir to the ion source. As their distance is long, a heater on a feed pipe was installed to prevent the vapor from condensing.
2. Charge stripping is performed by passing the ion beams through an electron stripping gas at a terminal in an accelerator. The efficiency of the charge stripping depends on pressure of the stripping gas which is affected by peripheral temperatures. A flow control valve of the stripper gas was wrapped in glass wool and aluminum foil as heat insulators to stabilize the pressure.
3. Beam line geometry from the exit of an accelerator tank to detectors was optimized by adjusting the position of the beam guide tubes.

Standard samples of NIST 4990c were measured. Relative standard deviations of ^{14}C/^{12}C ratios in 12 reputable measurements of 1 sample before and after the improvements were about 2% and 0.5%, respectively. Therefore the improvements enhanced stability of the ion beam.

1P26: Observation of local fields in zinc oxide with the ^{111}Cd probe

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Zinc Oxide (ZnO) is strongly expected as a semiconductor in a wide field of industries. The conductivity of semiconductors varies depending on the doping methods and the density of impurities. To control the conductivity of semiconductors, it is important to understand the functions of impurities in ZnO at an atomic scale. In our previous study, local fields in polycrystalline ZnO doped with 0.5 at.% indium (In) were measured by means of the time-differential perturbed angular correlation (TDPAC) method with the ^{111}Cd ($\leftarrow^{111}$In) probe, resulting in the detection of unexpected conduction-electron behavior at the probe sites. We infer that this phenomenon was caused by so-called “aftereffect’’ typically accompanying the electron capture decay of ^{111}In. In this work, we measured local fields in polycrystalline ZnO with the ^{111}Cd ($\leftarrow^{111m}$Cd) probe in order to investigate if the oscillatory structures of spectra are damped in the same way as the case for the ^{111}Cd ($\leftarrow^{111}$In) probe. When introducing the ^{111}Cd ($\leftarrow^{111m}$Cd) probe, a neutron capture product of ^{110}CdO, in the samples; it is essential to examine optimum condition for uniform diffusion of the probe. We therefore tried to synthesize samples at various annealing temperatures and chemical constituents. In this paper, showing parameter values such as the electric fields gradients, we discuss the aftereffect with respect to temperature dependence of spectral damping.

1P27: Synthesis and magnetic properties in Fe and Mn co-doped SnO_2 ferromagnetic semiconductors

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Ferromagnetic nanoparticles of diluted transition metal of both Fe and Mn co-doped into SnO_2 were synthesized by a sol-gel method. Fe and Mn co-doped SnO_2 enhanced the magnetization, which showed the maximum saturation magnetization value at 0.5% Mn and 1% Fe co-doping. With further increasing the amounts of Mn doping into 1% Fe-doped SnO_2 host, the magnetic-field dependence of magnetization showed the decrease in saturation magnetization. We found that the coexistence of Fe and Mn ions contributes to the increase of the magnetization. Secondary phase formation is excluded from the concentration dependences of magnetization and Mossbauer spectra. Broad magnetic relaxation peaks in Mossbauer spectra are related to the enhancement of the saturation magnetization. In the presentation, we discuss the co-doping effects into SnO_2 for the room-temperature ferromagnetism.

1P28: Temperature dependence of In-beam Mössbauer spectra of ^{57}Mn implanted into alminuum oxide

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Aluminum oxide (Al_2O_3) has been extensively investigated in materials science since its optical property is controllable by the nature and amount of impurities. In spite of the long history of studies, there still remain unresolved problems of the color changes arising from the presence of impurities. The objective of this work is thus to investigate the interacting nature of Fe ions implanted as impurities with the Al_2O_3 matrix by in-beam Mössbauer spectroscopy with the ^{57}Fe($\leftarrow^{57}$Mn) probe. Here, we have measured the emission Mössbauer spectra of ^{57}Mn implanted into Al_2O_3 of which temperature was controlled. The ^{57}Mn ions were produced as the projectile fragments of the high-energy ^{58}Fe beam. The sample was an Al_2O_3 single crystal plate (27 mm x 27 mm and 5 mm thickness). The emission Mössbauer spectra of ^{57}Mn implanted into Al_2O_3 were obtained at the temperature range from 92 K to room temperature. Details of temperature dependence of obtained Mössbauer spectra will be discussed.

1P29: Mössbauer absorption anomaly of gamma radiation via nuclear level anticrossing

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We have already reported a proof-of-principle experiment, demonstrating electromagnetically induced transparency (EIT) with gamma radiation for the Mössbauer effect using the level-crossing technique. In the extensive studies so far, we found that the new eigenstates might be observed.

We synthesized a single crystal of $FePSe_3$ by solid phase reaction in the vacuum-shielded tube (Vycol glass tube) including the mixture of stoichiometric amounts of Fe, P, Se at 840℃ for about 1 month. A powder sample of $FePSe_3$ was made by grinding the crystal.

From the analyses of a series of Mössbauer spectra taken at various temperatures, we determine that at 20K the states $|I=3/2, m=-3/2\rangle$ and $|I=3/2, m=1/2\rangle$ cross and are mixed.

In the Mössbauer spectrum taken with the powder sample, the absorption line that corresponds to $m_g=1/2 \leftrightarrow m_e=1/2$ transition was observed to be not a singlet but a doublet. So we conclusively considered that when two nuclear levels cross and are mixed, two newly generated eigenstates repelling each other could be observed experimentally under a certain situation.

1P30: Mössbauer spectroscopy of thermal synthesized kimzeyites.

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Kimzeyite belongs to garnet minerals and has a chemical form of $Ca_3(Zr,Ti)_2(Si,Al,Fe^{3+})_3O_{12}$. Garnet minerals are well known to show an excellent long-term stability of their structure. This property attracts an attention as potential storage media of radioactive nuclear waste. Kimzeyite is also considered as potential storage media because Hf can substitute Zr in kimzeyite. In order to use garnets as potential storage media, several composition of kimzeyites were synthesized at several temperatures under 1000 kg/cm^2 by using hydrothermal method. X-ray powder diffraction measurement confirmed that the obtained products were kimzeyite. In order to examine the state of Fe in obtained kimzeyite, ^{57}Fe Mössbauer spectra were measured. Fe occupies not only tetrahedral site but also octahedral site as in the case of yttrium iron garnet. Fe is found to be as Fe^{3+} ion in both tetrahedral and octahedral sites.

1P31: Spin-state change accompanied by structural change in the mixed crystals of assembled complexes bridged by 1,3-bis(4-pyridyl)propane

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$Fe(NCX)_2(bpp)_2$ (X=S, Se, BH_3; bpp=1,3-bis(4-pyridyl)propane has very rare structure which is 2D interpenetrated, while $Fe(NCX)_2(bpp)_2$・2(benzene) has 1D chain structure. By releasing benzene molecules, the host framework changed to 2D interpenetrated structure. The desorbed complex enclathrated benzene molecules again and host frame work returned to 1D chain structure. In the present study, we performed mixed metals experiment to know the mechanism of the structural change accompanied by sorption of benzene molecules. The reversible structural change of host framework was tried to explain by two factors: the crystal stability by making up the interstitial space and the conformer stability in coordinated bridging ligand.

1P32: The study of f1 configuration having small axial anisotropy

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As a member of light actinides, pentavalent or hexavalent uranium is characteristic of $[UO_2]^{n+}$ (n = 1 or 2) structure, so called "-yl structure", where two oxygen atoms are bonded to metal ion in axial symmetry. Magnetisms in pentavalent or hexavalent neptunyl or plutonyl have been explained by strong axial ligand field. Among the 5*f*-electronic states, uranyl(V) UO_2^+ is considered to be the most fundamental configuration ($5f^1$). UO_2^+ is thermodynamically unstable in the presence of protons due to a disproportionation reaction ($2UO_2^+ + 4H^+ \rightarrow UO_2^{2+} + U^{4+} + 2H_2O$), the study of the chemical properties of this system has been greatly constrained. On the uranium redox-flow battery proposed by our group, uranyl(V) is of great significance that is essential as the positive active material. We were measured at magnetic susceptibility of uranyl(V) complexes having $5f^1$ state. The room tempreture effective magneton moment (μ_{eff}) for β-diketonato complexes were determined to be 1.74, 1.66 and 1.58μ_B/U^{5+}. Note that the estimated moment values are below the theoretical value of 2.54 calculated for the free f^1 ion (U^{5+}) by in the ***L-S*** coupling scheme. The relatively smaller value of μ_{eff} at the room temperature is believed to be caused by the increased ligand field imposed on the uranium(V) center.

1P33: Stability constants of minor actinides and lanthanides with dithiocarbamate.

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(JAEA)

The Stability constants of trivalent minor actinides (An^{3+}; ^{241}Am and ^{244}Cm) and lanthanides (Ln^{3+}; Eu) with Ammonium pyrrolidinedithiocarbamate (APDC) in aqueous solution were investigated. The stability constants were obtained by back-extraction technique using di(2-ethylhexyl)phosphoric acid (HDEHP) as follows. Aqueous phase was acetic buffer solution from 0.02 to 0.10 mol/cm^3 containing various concentrations of APDC. Organic phase was toluene or benzene solution containing HDEHP and trace amounts of the extracted radionuclide such as ^{241}Am or ^{244}Cm. Eqvolumes of the aqueous phase and the organic phase were mixed in vial and were shaken 30 minutes at 298 *K*. The results show that minor actinides strongly interacted with dithiocarbamate. From those results, the obtained stability constants (β_n, n = 1, 2) for Am are $\log\beta_1$(Am) = 2.5 ± 0.3, $\log\beta_2$(Am) = 4.3 ± 0.2 at 298 *K*.

1P34: Development of an on-line detection apparatus using liquid scintillation detectors for analysis of spent nuclear fuel

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We have developed an on-line detection apparatus using a capillary electrophoresis system and liquid scintillation detectors for analysis of spent nuclear fuel. The detectors have been coupled to the capillary electrophoresis system by using a solvent extraction technique of flow injection analysis. In this work, the apparatus have been evaluated by using a mixture sample of lanthanides, U, Am, Cm, Cf, Cs, Sr, and Tc. From the results, it has been found that only the lanthanides and actinides are extracted into a liquid scintillation cocktail and introduced into the detectors in the condition of this experiment. The lanthanides and actinides have been able to be distinguished by lifetime discrimination of the emission from the scintillation cocktail. Moreover, the energy resolution of the detectors let us to distinguish the α-particles from the trivalent actinides and U.

1P35: Separation of U/Pu/Am for Safeguards Environmental Samples

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For accurate analyses of Pu in a Pu oxide / MOX particle, the sample should be chemically separated from Am and U. The mass of ^{241}Am which is daughter nuclide of ^{241}Pu and total mass of a polyatomic ion $^{238}U^1H$ induced from U overlap with those of ^{241}Pu and ^{239}Pu, respectively. These interferences lead to systematic error in the quantitative and isotopic analyses of Pu. In this work, two types of chemical separation technique of U, Pu, and Am were examined. One is an anion-exchange separation, and the other is an extraction separation with the TRU resin. Plutonium, U, and Nd which is analogous to Am in the chemical behaviors were successfully separated with the both an anion-exchange column combined with HCl media and a TRU extraction column combined with ascorbic acid for reduction of Pu. The decontamination factors between the elements of interest were more than 300 in case of the anion-exchange separation.

1P36: Study on dissipative behavior of radon gas using groundwater

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In the determination of radon in water sample, particular attention should be given to the radon loss from the sample. Estimation of the radon loss from the sample until the onset of measurement is very difficult. In addition, Rn loss process is associated with solubility of Rn gas. It is affected by solvent and temperature etc. In other word, they affect dissipative behavior of Rn gas. However, this behavior has not been clearly understood. In this work, in order to clarify that, laboratory experiments by using the spring water mentioned above were performed. For example, we observed the elapsed time variations of depth profiles of Rn loss using the 5L of sample water in a beaker. It was found that residual Rn in water tends to decrease with water depth. Concentrations of residual Rn in water were constant from the surface to bottom after still standing for 24 h. Because the water temperature was lower than the air temperature, convection was presumably generated in the beaker and this could be a cause of water depth dependence of Rn loss. We need to obtain further experimental data to clarify the dissipative process of Rn.

1P37: Well water analysis as a part of environmental monitoring

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Since 1989, we have performed environmental monitoring at Rokkasho village around Rokkasho reprocessing plant (RRP). The objects of the monitoring include well water, whose data were not frequently reported. We showed the concentration of ^{90}Sr in well water at site Obuchi-2 ranged 0.8 ~ 14 mBq/L and had increasing trend since 2003. It was approved that this trend was attributed to natural fluctuation because other artificial nuclides were not detected and specific activity of ^{90}Sr was constant since 2003. It was obvious that the ^{90}Sr originated from fallout of past nuclear tests, but this phenomenon attracts interest from geochemical viewpoint, so we conducted research to clarify the mechanism. We compared the concentration of ^{90}Sr, stable Sr, electric conductivity etc in the water and soil samples taken from nearby points. Because the concentration of ^{90}Sr in well water had good correlation with stable Sr, electric conductivity and other ions, we assumed ^{90}Sr fluctuated along with other soluble substances. Because the increasing trend started before the active test of RRP (since March 2006), and large scale land reclamation was conducted in upstream part until 2003, we assumed the behavior of ^{90}Sr and other substances was influenced by change of hydrological condition of surrounding environment.

1P38: The improvement of detection limit for Cs-137 in soil using Ge detector with AMP procedure

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In some case, many of gamma ray peaks from natural radionuclides such as K-40, U-series, Th-series, disturb the detection of Cs-137 in soil with Ge detector. We applied the ammonium molybdophosphase (AMP) precipitation method to remove these natural radionuclides. The recovery of Cs was obtained by AAS measurement of the dilution that dissolved the fraction of AMP precipitation in NaOH. The gamma spectrum of AMP shows the peak of Cs-137 more definitive, and significant improvement of detection limit. The results of AMP precipitation method were consistent with direct measurement.

1P39: Determination of long-lived radionuclides ^{129}I and ^{36}Cl in soils from Futaba area, Fukushima prefecture

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^{129}I (half-life: 1.57×10^7 yr) and ^{36}Cl (half-life: 3.01×10^5 yr) are important long-lived radionuclides used as environmental tracers. They are produced both naturally and through human nuclear activities. The majority of ^{129}I has previously been accumulated in surface soils by global fallout, whereas the behavior of ^{36}Cl has not been understood in detail. The aims of this work are to determine ^{129}I and ^{36}Cl activities in soils, and to compare the two nuclides on their environmental distributions. Isotopic ratios of ^{129}I/I and ^{36}Cl/Cl were determined by accelerator mass spectrometry (AMS). In addition, to investigate soils in more detail, we determined organic matter content in soils by loss on ignition (LOI) and ^{137}Cs concentration in soils by an HPGe detector. Samples were collected at 8 points from Futaba area, Fukushima prefecture. We investigated the distribution of long-lived radionuclides in surface soils (0~10 cm) at all points and 1 meter depth profile at one point.

1P40: Metal binding characterization of hemocyanin purified from liver of squid

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One of the artificial radionuclides, silver-108m, is still detected in liver sample of flying squid caught around Japan. We purified a silver binding protein from liver of squid and identified it as respiration pigment hemocyanin. Here we show the metal binding characterization of hemocyanin with 9 radioactive elements. Silver showed good binding (0.34 μg-Ag/mg-protein (p)) to hemocyanin compared to bovine serum albumin (BSA), which was used for negative control. The amount of silver bond to hemocyanin was not apparently decreased by addition of copper ion. The result suggested that the binding site of silver was not the center of copper binding site. Although Cadmium was bond to henocyanin at a high rate (3.9 μg-Cd/mg-p), the same amount of cadmium was bond to BSA suggesting that the cadmium binding is not specific. Strontium, calcium, cobalt, manganese and zinc were also bond to hemocyanin. The bindings of strontium and cobalt were not inhibited by addition of copper ion. On the other hand, the bindings of the other three metal elements were competitively inhibited by copper suggesting that these metals may be bond to copper binding site. Cesium and technetium were not bond to hemocyanin.

1P41: Role of Radiochemistry in the Comprehensive Nuclear-Test-Ban Treaty

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Verification of nuclear explosion under the Comprehensive Nuclear-Test-Ban-Treaty (CTBT) is carried out by remote monitoring of geophysical signals and radionuclides through international monitoring system (IMS) and on-site inspection (OSI). Radioxenon nuclides including ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe, and radionuclide particles as well as geophysical signal are automatically monitored by the IMS. Gamma-ray monitoring and radionuclide analysis are important inspection technologies for OSI. Both gamma-ray dose and energy spectrum are monitored from air and on the ground to narrow down the inspection area. Radionuclide analysis of environmental samples is performed to determine the event as a nuclear explosion. In addition to radionuclide analysis in the IMS and the OSI, radiochemists are requested to consult the national authorities for an explanation of scientific data to make national decision pursuant to the Treaty. Role of and anticipation to radiochemistry in the CTBT are presented.

1P42: Annual variation of ^{14}C isotope ratio in plant samples (tree rings or rice) by AMS

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^{14}C ($T_{1/2}$; 5730yr) is produced in the atmosphere by the reaction of cosmic ray with nitrogen. Plants remove $^{14}CO_2$ from the atmosphere together with $^{12,13}CO_2$ by photosynthesis. Tree rings or rice grains have ^{14}C/^{12}C ratios related to that in the atmosphere. The ratios in the atmosphere are affected by solar activity. We have determined ^{14}C/^{12}C ratios in tree rings of Yaku-cedar from AC1000 to 1100. As a result, we observed a peak of the ratios around AC1150, suggesting the solar activity would be weaken in this period.

We also measured ^{14}C/^{12}C ratios in grains of rice collected from 1950 to 2009, to estimate the variation of atmospheric ^{14}C levels which were influenced by nuclear weapons testing. The highest peak of ^{14}C/^{12}C ratio was found around 1963 and then it decreased gradually.

Tree rings of pine collected from the vicinity of Chernobyl NPP to assess the release of ^{14}C at the accident, which occurred in 1986. A peak of ^{14}C/^{12}C ratio clearly observed in the tree ring of 1986. In order to know the distribution of ^{14}C in this tree ring, we will determine both early and late wood.

1P43: Distribution of Pu isotopes and Am-241 concentration in the Eastern Pacific

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Geographical and depth distributions of anthropogenic radionuclides of Pu-238, 239, 240, and Am-241 were studied in the Eastern Pacific in 2003. From a result of analyses of Pu isotopes and Am-241 in seawater columns and sediment columns, it was found that the Pu-239, 240, Pu-238 and Am-241 inventories in water columns are 0.78-5.11 Bq/cm^2, 0.62-1.95 Bq/cm^2, and 5.32-21.7 Bq/cm^2, respectively, while the Pu-239, 240, and Pu-238 inventories in sediment columns are 0.34-1.26 Bq/cm^2 and 0.01-005 Bq/cm^2, respectively. The value of Pu-238/Pu-239, 240 activity ratios of water-column coincided with that of sediment-column in stations of North Pacific and Equatorial Pacific. They agree with that of global fallout. Therefore, the Pu isotopes in these sea areas are considered to originate from global fallout. On the other hand, the Pu-238/Pu-239,240 activity ratios in seawater of South Pacific indicate a high value of 0.05-0.11, so that it might be affected by the release of Pu-238 from the SNAP-9A artificial satellite accident.

1P44: Distribution of Pu isotopes in a size-fractionated surface soil sample in northern China

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Two years ago, in the 52th Radiochemistry symposium, we reported the Pu activity level and the isotopic characteristics in soils in Gansu Province, northwest China. In this work, we present a recent investigation on the distribution of Pu isotopes in a size-fractionated surface soil sample collected in a grassland in Jining, the Inner Mongolia, in northeastern China. Soil grain size fractions were determined with sieves. This surface soil consisted of sand (> 53 μm, 89.6%), with silt and clay (< 53 μm) contributing ca. 11%. Plutonium isotopes were analyzed using SF-ICP-MS after digestion with HNO_3-$HClO_4$-HF and ion-exchange chromatographic separation/purification. The activities of $^{239+240}$Pu ranged from 0.10 to 0.52 mBq/g, and decreased nearly exponentially with increasing particle size of the soil due to a decreasing surface area. The activity of $^{239+240}$Pu in the finest particle (<25μm) which can be transported to long-distance in the dust event, was ca. 5 times higher than that in the coarse particles (600-1000 μm). In contrast to the significant variation of $^{239+240}$Pu activity, the atom ratios of ^{240}Pu/^{239}Pu did not show any variation, they ranged from 0.169 to 0.200, with a mass-weighted mean of 0.185 ± 0.025, indicating that Pu originated from global fallout. Although more studies are needed, the results obtained in this study provided further evidence to support the hypothesis that the temporal change of $^{239+240}$Pu 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.

1P45: QUANTITATIVE ANALYSIS OF ENVIRONMENTAL RADIONUCLIDES IN IMPORTED FOOD- ON THE IMPORTED FOOD FROM CHINA

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The food imported from China and its neighboring countries were heated to 450℃ with an electric furnace and were made ashes, then measured with a Ge spectrometer. Based on the counting rate (cps) of the full energy peak, the concentration of the γ-emitting radionuclides (Bq/kg) was determined. From the ash sample, which was heated again to 500℃, ^{90}Sr was separated by the oxalate method. After secular equilibrium between ^{90}Sr and ^{90}Y was attained, the samples were measured with a liquid scintillation spectrometer using the modified integral counting method. The concentration of radionuclide (Bq/kg) were: tea leaves; ^{137}Cs(0.17±0.02 ～ 1.08±0.06), ^{90}Sr(3.74±0.55～12.6±2.8), ^{40}K(450～2100), the frozen food; ^{137}Cs(ND～0.14±0.02）,

^{90}Sr(ND, except for one sample), dried sea fish; ^{137}Cs(0.055±0.014～0.13±0.02). The concentration of ^{40}K, ^{226}Ra, ^{228}Ra in the frozen food were lower than average concentration of these nuclides in the soil. The results obtained here do not seem to include an urgent problem of hygiene. However, we have to emphasize that ^{90}Sr and ^{137}Cs must be removed from the food and the feed of livestock, because these nuclides are condensed by the food chain, and accumulated in a human body.

1P46: Measurement of tritium concentrations in rain and airmass backtrajectory analysis

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Tritium is a radioactive isotope of hydrogen with 12.33 years half-life and is produced naturally in the upper atmosphere by interaction with secondary cosmic radiation. A large quantity of tritium will be used as a fuel at nuclear fusion reactor that would be a potential source of environmental tritium in future. Tritium concentrations in rain water in Fukuoka city and Toki city were determined by combination of electrolytic enrichment and liquid scintillation counting. The average of tritium concentrations observed in rain in Fukuoka and Toki were 0.79 ± 0.33 Bq/l (2005/5~2009/7) and 0.38 ± 0.14 Bq/l (2007/6~2010/3) respectively. A cyclic seasonal variation is observed in both cities: high in winter/spring and low in summer/autumn. Tritium concentration in rain in Toki has however showing a smaller variation compare with that in Fukuoka. The back trajectory analysis of airmass suggests that the rain clouds arrived at Fukuoka from the Eurasia continent give rain with higher tritium concentration than those from the Pacific Ocean. The seasonal variation observed in tritium concentration is concluded to be originated from migration pathway of airmass which is strongly dependent upon meteorological factors. In symposium, we also report the result of the back trajectory for Toki city and discuss the difference of tritium concentrations in both cities.

1P47: Seasonal variations of ^{228}Ra, ^{226}Ra, and ^{228}Th activities in surface water on the east side of the East China Sea and their implications

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In the surface of the East China Sea (ECS), the Kuroshio water (KW) and the continental shelf water (CSW) are mixed, then the mixed water injects into the Japan Sea as Tsushima current. To understand material transport from ECS to the Japan Sea, the mixing mechanism of two water mass should be elucidated. The surface sea water samples were collected at 5 sites on the east side of ECS in 2-3 months interval during the *Yoko Maru* and *Soyo Maru* expeditions, and ^{226}Ra, ^{228}Ra, and ^{228}Th activities were measured using low-background γ-spectrometry. The ^{228}Ra/^{226}Ra and ^{228}Th/^{228}Ra activity ratios among the samples exhibited notable seasonal variation (^{228}Ra/^{226}Ra = 0.4-2.7; ^{228}Th/^{228}Ra = 0.1-0.4) accompanying the changes of salinity (31.3-34.5). These changes are hypothesized to cause the change by altering the mixing ratio of ^{228}Ra-rich and high-particle CSW within the ECS. However, the variation of ^{228}Th/^{228}Ra ratio does not closely agree with the mixing ratio of CSW (^{228}Ra/^{226}Ra and salinity). In this study, we compare the seasonal variation of ^{228}Ra/^{226}Ra and ^{228}Th/^{228}Ra activity ratios in surface water and discuss the transport of water mass and particle reactive materials in ESC.

1P48: A new system of Pb-210 measurement for sedimentation rates at the Geological Survey of Japan

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We have measured the sedimentation rates of lakes and sea bottom sediments by use of the well-type Ge semi-conductor detectors at the Geological Survey of Japan, AIST. Now we renewed the measurement system because they became old and sometimes had trouble with the detector. In this presentation we show the details of the new measurement system and some results of sedimentation rates at the Huanghe estuary that were measured in this system. The system is composed of the well-type Ge detector (Ortec GLW-120230-S), J type cryostat and shielding (Pb 115mm, Fe 48mm, Cd 2mm, Cu 20mm, acryl 5mm). The continuous background level of gamma spectrum decreased when the J type cryostat was used rather than the vertical type cryostat was. The oxygen free copper (OFC) materials also lowered the background level (continuous and peak) than the Al materials. The bottom sediments were taken at the Huanghe estuary in 2009 using gravity corer and the activities of lead-210, lead-214 and cesium-137 were measured.

1P49: Estimation on transport of riverine suspended solids in Satoyama area using ^{137}Cs and ^{210}Pb$_{ex}$ as geochemical tracers

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Investigation of sediment transport in river systems is important to make protection of food production and sediment disaster. In this study, riverine suspended solids (SS) have collected in the Kumaki River, which runs through abandoned Satoyama area in Noto Peninsula. To estimate the main source of SS, surface soil samples were also collected from managed and unmanaged forest, and paddy field. The samples were analyzed for ^{137}Cs and ^{210}Pb$_{ex}$ by gamma spectrometry. The low ^{137}Cs /^{210}Pb$_{ex}$ activity ratios are found in surface samples of abandoned paddy rice field associated with accumulation of ^{210}Pb$_{ex}$ relative to ^{137}Cs. The low ratio of ^{137}Cs /^{210}Pb$_{ex}$ is also found in surface soil samples of unmanaged forest because heavy rain erodes surface soil. The river bottom sediments show relatively low ^{137}Cs /^{210}Pb$_{ex}$ activity ratio in the watershed area that runs through unmanaged forest, paddy rice field, and abandoned paddy rice field. These results indicate that in the Kumaki basin, particle transport processes are controlled by surface soil erosion from the forest area and paddy field by washout of heavy rain.

1P50: Determination of trace elements and transformation of proteins in pancreas of zinc-deficient mice

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Concentrations of trace elements and proteins in pancreas of Zn-deficient and control mice were investigated. Eight weeks 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 a week, their pancreases were removed and every four pancreases of each group were together homogenized in a Tris buffer. Then, a cytosol fraction was separated from the homogenate by centrifugal separation method. SDS-PAGE and 2D electrophoresis were carried out. Proteins in the cytosolic fraction were also separated into twenty fractions by gel-filtration chromatography. After that, SDS-PAGE was performed for each fraction. New bands were found on the gel for a few fractions of Zn-deficient mice. Bradford protein assay and atomic absorption spectroscopy were also performed for each fraction. For concentration of protein in all fractions, there were no significant differences between two groups. On the other hand, concentrations of zinc in several fractions of Zn-deficient group were decreased. These results indicate that some zinc-bound proteins may exist for apo-proteins.

1P51: Effects of zinc deficiency on behavior of trace elements and change of metalloprotein in testes in mice

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Zinc is one of the most important essential trace elements. Zinc deficiency leads to skin injury, alopecia, growth retardation, loss of sense and so on. The production of testosterone and spermatogenesis are also affected by zinc deficiency. Eight weeks old male mice of ICR strain were divided two groups. One is Zn-def. group fed with Zn-def. diet, and other is control group fed with control diet. After a week, their testes were removed and every four testes of each group were together homogenized in a Tris buffer. Then, a cytosol fraction was separated from homogenate by centrifugal separation method. SDS-PAGE and two-dimensional electrophoresis were carried out. When the positions and the number of the protein spots in gel two-dimensionaly developed the cytosolic fraction of Zn-def. mice were compared with those of control ones, significant differences were found between them.

1P52: Evaluation for olfactory transport mechanism of intranasally administered ^{201}Tl

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Our laboratory has found that the migration of a portion of heavy-metal ions such as $^{201}Tl^+$ can access to the brain bypassing the Blood-Brain Barrier via olfactory neurons. This phenomenon is referred to as an olfactory transport, clinical development of olfactory functional diagnosis using this pathway is now undergoing. However, its transport mechanism has not proven yet. In this study, for elucidation of the mechanism, we studied the intranasal delivery to the brain by using ^{201}TlCl and ^{99m}Tc-MIBI, both of them were used for myocardial perfusion imaging. Moreover, we weighed the inhibition effect for the brain of three inhibitors, ouabain, cholchicine, and furosemide, by using double tracers, ^{201}Tl and ^{54}Mn. Anesthetized ICR male mice were intranasally administered the prepared solutions mixed with ^{201}Tl and ^{99m}Tc-MIBI or ^{54}Mn into the right nostril. In this experiment, mice were pretreated with intranasal administered inhibitor. The animals were sacrificed 1, 3, 6, 12, 24 hours, and 3, and 7days after double tracer administration. While uptake of ^{201}Tl into the olfactory bulb was gradually increased with time, uptake of ^{99m}Tc-MIBI was not observed. Both ouabain and cholchicine treated group, uptake of ^{201}Tl into the brain was not affected by their inhibitors.

1P53: Synthesis of ^{186,188}Re-MAG3 and ^{186,188}Re-[(CO)$_3$ (H$_2$O)$_3$]$^+$

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Both ^{186}Re (β ray$_{max}$ 1.08 MeV) and ^{188}Re (β ray$_{max}$ 2.12 MeV) are expected as therapeutic nuclide in the cancer treatment and the pain easing. Synthesis of ^{186,188}Re-MAG3 (mercaptoacetyltriglycine) complex and ^{186,188}Re-[(CO)$_3$ (H$_2$O)$_3$]$^+$ (tricarbonyl) complex were examined under various conditions

1P54: Treatment for osteoblastic metastasis in rats using α-particle emitting ^{227}Th-EDTMP

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Most patients with advanced breast, prostate, or lung carcinoma develop metastases. Especially, bone metastases cause severe pain, so there have been many attempts such as surgical invention, chemotherapy, external radiation therapy, and so on. However, many of these treatments are limited in their efficacy or duration and have significant side effects.

Due to the high LET and short range of α-particles in tissue, α-emitting nuclides are promising for the treatment of bone metastases. Reduction of bone marrow exposure can be achieved due to the short range of α-particles. In this study, we evaluated the therapeutic effect of α-emitting ^{227}Th-EDTMP for osteoblastic metastasis in rats.

Copenhagen strain rats were inoculated with $3x10^3$ cells of syngeneic prostate cancer cell line MAT-LyLu into the right tibia. One week after cell inoculation, 250kBq/kg b.w. of ^{227}Th-EDTMP was injected into rats via tail vein. During 4 weeks after cell inoculation, antitumor effect and pain palliation effect were examined using tumor volume measurement and von Frey filament test, respectively.

The von Frey filament test showed bone pain palliation 2 weeks after cell inoculation. Tumor growth were inhibited 17 days after cell inoculation, however there were no tumor growth inhibition on 21 days after cell inoculation.

1P55: Reduction of the cosmic ray components of the Ge detector utilizing the thin plastic scintillators for anticoincidence

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The contribution of cosmic ray components on background of Ge detector is reduced by means of an active anti-coincidence detector system. In order to insert into lead shield and cover 2π geometry of Ge detector, five thin plastic scitillators (PSs) (180 mm x 180 mm, and 3 mm in thickness; ELJEN TECHNOLOGY; EJ-200) 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) through five clear fibers and photo-couplers has been developed. 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 re-tested. This thin PSs system has sufficiently thickness to detect cosmic ray components and expects to reduce more than 50% of those induced background components.

1P56: Correlation between the particle size distribution of radioactive aerozols and their half-lives -Based on the experimental results in 120 GeV proton target station-

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In Anti-proton target station (AP0) in Fermi National Accelerator Laboratory, the Inconel target is irradiated by ultra-high energy protons at 120 GeV. In terms of external and internal exposure, it is important to understand the behavior of radioactive aerosols in an accelerator target room. In this work, we separated radioactive aerosols collected from the target room of AP0 into several samples according to particle size by using the impactor method, and obtained particle size distribution of radioactive aerosols associated with corresponding radio-nuclides through measuring activity of various radio-nuclides in those samples by gamma-ray spectrometry. Consequently, it was found that the particle size distribution of radioactive aerosols tends to depend on their half-lives of their corresponding radio-nuclides. Based on the results, the behavior and the formation mechanism of radioactive aerosols in the target room will be considered in detail. Additionally, to obtain the information about smaller size of radioactive aerosols than those separated by the impactor method we collected radioactive aerosols in the same place as noted above using the graded screen array (GSA) method. We also intend to present the results obtained by GSA method.

1P57: Simple determination method for ^{99}Tc in radioactive waste arising from research facilities

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On disposal of radioactive waste packages, an evaluation of radioactivity inventories in the packages is required. Technetium-99 is one of the important nuclides from the viewpoint of safety assessment for disposal of radioactive wastes, because of its very long half life. In the present study, a simple ^{99}Tc determination method was developed for surface contaminated metal wastes. First, pretreatment conditions for removing ^{99}Tc from the surface of metal were studied. It was found that recovery of ^{99}Tc by soaking with acids was higher than that by alkali fusion with sodium hydroxide. More than 90% of ^{99}Tc could be recovered from the sample solution containing large amounts of iron (>0.5g) using a solid phase extraction resin (TEVA-Resin). In the separation using the resin, the recovery of ^{99}Tc was good agreement with that of Re, which was added as a tracer. Finally, the simple ^{99}Tc determination method was successfully applied to actual radioactive metal waste samples arising from Japan Power Demonstration Reactor of the JAEA.

2A01: Electronic structure of chalcogenide–capped octahedral hexatechnetium complexes

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The HOMO/LUMO levels, the frequency of the C≡N stretching bands, and the electronic transition energies in $[Tc_6Q_8(CN)_6]^{4-}$ (Q = S or Se) were determined by DFT calculations. The LUMO is localized on the technetium atoms in both hexatechnetium complexes. The HOMOs are localized on the Tc_6Q_8 core. The plot of the energy level of HOMO in $[M_6Q_8(CN)_6]^{4-}$ (M = Tc, Re; Q = S, Se) against the redox potential of the M_6(23e/24e) process exhibits a good linear correlation. The HOMO-LUMO energy gap of the hexatechnetium complexes is approximately 0.6 eV (4800 cm^{-1}) smaller than that of $[Re_6Q_8(CN)_6]^{4-}$.

Electronic singlet transitions from the HOMO regions to the LUMO regions were also calculated using TD-DFT method. The TD-DFT calculations for both sulfide- and selenide-capped hexatechnetium complexes indicate that the substantially allowed singlet transitions in the range of 400-500 nm are ascribed to the Tc_6Q_8 core-centered. The longer wavelength shift of the selenide-capped complex at the lowest energy electronic transition compared to that of the sulfide-capped complex in the hexatechnetium complex shows the same trend in the experimental UV-vis data in acetonitrile.

2A02: Synthesis and Redox Properties of Bis(salophen)uranium Type Complexes

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The substituent effect to the redox properties of uranium complexes produced by the reaction of UCl4 with bis-salophen type ligands ($H_2(R_1/R_2)$ = *N,N'*-(4,5-R_1-1,2-phenylene)bis(5-R_2-2-hydroxybenzylidenimine), (R_1/R_2) = Cl/Cl (**1**), Cl/OMe (**2**) were studied. In **2**, two ligands coordinate to a uranium atom to give eight-coordinate structure. The two ligands are oriented head-to-tail conformation. This may be due to the steric hindrance of the substituent groups at R_1 and R_2 positions. The U-O (av. 2.24(4) Å) and U-N (av. 2.62(4) Å) distances are resemble those of the congeners. The redox potentials of **1** and **2** were measured by cyclic voltammetry in CH_2Cl_2. The plot of the redox potentials versus *Hammett constants* (para effect) shows linear correlations. This indicates that the electron donating/withdrawing ability of the donor atoms directly affects to the redox potential of the actinide center.

2A03: Selective electrocatalytic reduction of Np(V) to Np(IV) at a flow-through electrode

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Neptunium(V) ion is reduced by electrolysis through two reduction paths, i.e. the chemical reaction with Np(III) produced from Np(IV) by electrode reaction and the electrocatalytic reduction by adsorbed hydrogen atom on platinum electrode surface. Based on the electrocatalytic reduction, development of rapid and selective electrolytic reduction of Np(V) to Np(IV) is expected. A flow-through electrode of glassy carbon (GC) fiber platinized by electrodeposition was examined in the present study. Although Np(V) was reduced to Np(III) at the much negative potential at a flow-through electrode of normal GC, stepwise reductions of Np(V) to Np(IV) and Np(IV) to Np(III) were observed on the potential-current relation of electrolysis at platinized GC electrode. The overpotential of reduction of Np(V) to Np(IV) was decreased. This electrolysis method enables us to prepare Np(IV) rapidly without producing Np(III). Np(V) in the mixture solution of Np and U was also reduced to Np(IV) selectively, not accompanied with reduction of U(VI).

2A04: The new methods for preparing trivalent uranium and its structure and magnetis

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The uranium(III) compounds have attracted large interests in the coordination chemistry, catalysis, magnetism and separation chemistry. Uranium(III) compounds are prepared under reducing condition because of its instability against rapid oxidation. In the preparation of uranium(III) organometallic complexes, halides such as iodide and chloride have been used as the important starting material. Uranium(III) triiodides such as UI_3 and $UI_3(thf)_4$ (thf = tetrahydrofuran), which has been prepared for uranium metal, have played a major role in synthesis of uranium(III) organometallic complexes. We have developed a new method to prepare $UCl_3(thf)_{1.5}$ combining mercury-cathode electroreduction and the vacuum drying. $UCl_3(thf)_{1.5}$ was measured the temperature-dependent magnetic susceptibility and the effective magnetic moment was determined to be 3.58 μ_B, which is near to the expected value for f^3

($3.64\ \mu_B/U^{3+}$). And crystal structure was determined at pyridine adduct of uranium (III) chloride complex. This compound is convenient for use as starting material because of easy dissolution into various polar solvents. We report a simple and easy preparation method of uranium(III) chlorides from uranium oxide. And the structure and the magnetism of the prepared uranium(III) chloride with solvent adduct were studied in solid state.

2A05: Solvent extraction of heavy actinides (III) with HDEHP

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The extraction behavior of trivalent actinides (Am, Cm, Cf, Es, and Fm) with di(2-ethylhexyl)phosphoric acid, HDEHP, into benzene has been studied together with a series of lanthanides.

Fermium-250 ($T_{1/2}$ = 30 min) was produced in the ^{238}U(^{16}O,4n) reaction using the AVF cyclotron of RCNP, Osaka University. The reaction products recoiling out of the target were transported through a Teflon capillary to a chemistry laboratory. The transported products were deposited on a Naflon sheet for 30 min and were then dissolved with 0.2 mL of 0.1 M HNO_3. The aqueous phase was mixed with 0.2 mL of 0.1-0.3 M HDEHP-benzene solution and was then shaken for 10 min. After phase separation by centrifugation, 0.16 mL aliquots of the two phases pippeted were separately evaporated to dryness on 2 Ta dishes, which were subjected to α-particle spectrometry with Si PIN photodiode detectors. The extraction constants K_{ex} were determined by the slope analysis for the variation of the distribution ratio as a function of the concentration of HDEHP. The K_{ex} values of lanthanides, Am, Cm, Cf, and Es were also determined by the batch-wise extraction experiments using non-radioactive lanthanides and radiotracer of ^{241}Am, ^{243}Cm, ^{249}Cf, and ^{253}Es. We found that the extraction constant of Fm is smaller than that of Dy in spite of the similar ionic radii, while the K_{ex} values of other actinides are almost the same as those of each lanthanide with a similar ionic radius.

2A06: Challenge for establishing a simplified separation process of trivalent minor actinides using novel R-BTP adsorbents

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In order to directly separate trivalent minor actinides (MA: Am, Cm) from fission products (FP) containing rare earths (RE) in high level radioactive liquid waste (HLLW), the authors have been challenging for establishing a simplified MA separation process by extraction chromatography. We have attempted a new type of nitrogen-donor ligand, R-BTP (BTP: 2,6-bis (5,6-dialkyl-1,2,4-triazin-3-yl)pyridine, R: alkyl group) as an extractant because it shows high extraction selectivity for Am(III) over RE(III). It was reported that the R-BTP extractants showed different properties such as adsorbability and stability by having different alkyl groups. Therefore, novel R-BTP extraction adsorbents were prepared by impregnating appropriate R-BTP ligands into the porous silica/polymer composite support (SiO_2-P particles).

This presentation deals with *i)* adsorption and desorption properties of Am and FP in simulated HLLW solution containing HNO_3 onto the *iso*hexyl-BTP adsorbent, *ii)* hydrolytic and radiolytic stabilities of the adsorbents were examined, and *iii)* other type of R-BTP adsorbents were also prepared and investigated.

2A07: Basic research of spent fuel reprocessing process based on sulfurization: (1) sulfurization of tracer doped sample

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For the recovery of fissile materials from spent nuclear fuel, we have proposed a novel reprocessing process based on selective sulfurization of fission products (FPs). The key concept of this process is utilization of unique chemical property of carbon disulfide (CS_2), i.e., it works as a reductant for U_3O_8 but works as a sulfurizing agent for trivalent actinides and lanthanides. In this study, actinide tracers (^{236}Pu, ^{239}Np, ^{241}Am) and FP traces (^{152}Eu, ^{137}Cs, ^{90}Sr) were doped to U_3O_8 to prepare a simulated spent fuel sample. This RI tracer doped sample was sulfurized at a range of temperature 300 to 550 ^{o}C by flowing CS_2 gas for 1 hour. After sulfurization, the sample was immersed into 1M nitric acid solution and shaken for 1 hr at 50 ^{o}C. Then, the solution was separated by suction filtration to determine the relative concentration of each elements in the solution by alpha- and gamma-ray spectrometry. The separation ratios of FPs from U and Pu and the applicability of this procedure for the spent fuel reprocessing are discussed in the presentation.

2A08: A newly developed rhenium/technetium-99m separation method from molybdenum matrix

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It is necessary to develop new separation methods for technetium-99m (^{99m}Tc) from molybdenum (Mo) to use domestically produced ^{99m}Tc in radiopharmaceuticals. National Institute of Radiological Sciences plans to develop its own method for ^{99m}Tc supply using ^{100}Mo (p, 2n) ^{99m}Tc reaction. To separate ^{99m}Tc from Mo, a sequential use of Tc-selective extraction chromatographic resin (TEVA resin, Eichrom) and anion exchange resin (Dowex 1X4, Muromachi Kagaku Kogyo) was investigated. Rhenium (Re) was used as a chemical analogue of ^{99m}Tc. Our goal is to obtain a high Mo recovery for reuse, minimize Mo concentration of < 1 μg/mL in the final Re/Tc fraction, and recover Re/Tc into HCl solution. In the present procedure Mo powder was dissolved in a mixture of H_2O_2 and NH_4OH, and then Re was added. After extraction of Re on TEVA resin from the Mo solution, Re was recovered with 8N HNO_3 at almost 100% recovery. Mo remained in the Re fraction was removed using Dowex 1X4 and finally Re was eluted with 6N HCl solution. Mo concentration was low enough (< 1 μg/mL) and Re recovery was about 60-70%. When we used the method for ^{99m}Tc, total recovery of 85-88% was achieved.

2A09: Productions of ^{52}Fe and ^{62}Zn and Synthesis of iron- and zinc-hematoporphyrin complexes for PET imaging

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Iron-52- and zinc-62-labeled hematoporphyrin would be promising reagent for the cancer diagnosis because it is known that porphyrin-based molecules accumulate in the cancer cells. In this work, we studied the rapid separation methods for ^{52}Fe and ^{62}Zn tracers from the target materials and the synthesis of iron- and zinc-labeled hematoporphyrin complexes in a tracer scale for PET imaging. The isotopes ^{52}Fe and ^{62}Zn were produced in the ^{nat}Cr (^{3}He,*xn*) ^{52}Fe and ^{nat}Ni (α,*xn*) ^{62}Zn reactions, respectively, using thce AVF cyclotron at Research Center for Nuclear Physics, Osaka University. The Cr_2O_3 and Ni powders were mixed with KCl powder and used as the target materials. The reaction products attached to KCl were dissolved in aqueous HCl solutions of pH 3 and separated from the insoluble target materials, namely Cr_2O_3 and Ni. The amounts of radioactivities of ^{52}Fe and ^{62}Zn separated from the targets were measured by γ-spectrometry using Ge detectors. Zinc-65-labeled hematoporphyrin was successfully synthesized in a tracer scale. We succeeded in rapid synthesis of the iron-hematoporphyrin complex by using microwave. The synthesis of ^{55}Fe-labeled hematoporphyrin will be performed in a tracer scale.

2A10: Study of the ^{188}W-^{188}Re generator using PZC

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Re-188 (β ray$_{max}$ 2.12 MeV) is expected as therapeutic nuclide in the cancer treatment and the pain easing. Poly Zirconium Compound (PZC) has the adsorption capacity of the tungsten tens of times or more the alumina. The basic experiment was done for preparation the small size and practical ^{188}W-^{188}Re generator using PZC with different synthesis conditions. Adsorption yield of ^{188}W to PZC ranged from 40% to 100% by heating of 90℃ and 180 minutes.

2B01: Studies on hot atom chemical behavior of energetic ions in solids(XXIII)

-Study of Retention Behavior of Hydrogen Isotopes in Boron Film Deposited on SS-316 –

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Boronization has been applied as one of the most effective techniques for first wall conditioning in Large Helical Device (LHD). Boron is easily bound to impurities and the impurity-contained boron film will be formed. It is considered that energetic tritium from plasma will be implanted and retain into boron film. It is thought that chemical state of oxygen would influence the retention behavior of hydrogen isotopes due to the stable hydrogen trapping by oxygen.. In the present study, the influence of oxide layer on deuterium retention was studied using two different substrate of silicon and SS-316.

The boron films were prepared by the plasma CVD apparatus on the SS-316 and the silicon substrates, and deuterium ions (D_2^+) were implanted into the films with ion fluencies of 1.0×10^{22} $D^+ m^{-2}$ The chemical states and the atomic ratio of boron films were evaluated by XPS and the deuterium desorption behavior was revealed by TDS.

The major chemical state of boron on silicon is B-O bond, while that on SS316 is B_2O_3.In D_2 TDS spectra for SS316 substrates, three D_2 desorption stages were located around 500 K, 630K and 780 K, indicating that these stages were desorption processes of deuterium trapped as B-D-B, B-D and B-O-D bonds. The deuterium retention for SS316 is less than for silicon, suggesting that deuterium desorption behavior depend on changing chemical state of boron film for oxygen layer.

2B02: Studies on hot atom chemical behavior of energetic ions in solids(XXIV) ~Elucidation of chemical behavior of deuterium in tungsten under simultaneous energetic triple ion (C^+, D_2^+ and He^+) implantation

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In D-T fusion reactors, the combination usage of tungsten and CFC has been considered as plasma facing materials especially for the divertor. During plasma operation, it is anticipated that carbon will be sputtered by energetic particles and implanted into the surface of tungsten with energetic hydrogen isotopes and helium from plasma. Therefore, it is predicted that simultaneous implantation of carbon and helium affects on hydrogen isotopes retention including tritium. In the view point of tritium recycling, elucidation of tritium retention behavior in tungsten under the simultaneous ion implantation is one of the most important issue. In this study, D_2^+, C^+ and He^+ were implanted into tungsten simultaneously and the deuterium retention behavior in tungsten was investigated by means of TDS and XPS. Comparing the D_2 TDS spectrum for D_2^+-C^+-He^+ implanted tungsten to those for D_2^+-C^+ and D_2^+-He^+ implanted one, it was found that D_2 TDS spectrum for D_2^+-C^+-He^+ implanted tungsten showed almost the same D_2 desorption stage as that for D_2^+-He^+ implanted one at 400-600K, and the D_2 desorption stages around 800 K observed in D_2 TDS spectrum for D_2^+-C^+ implanted tungsten were not appeared. These facts indicate that implanted He^+ prevents the trapping of D as C-D bond and by irradiation defects.

2B03: Studies on hot atom chemical behavior of energetic ions in solids (XXII)- Annihilation behavior of irradiation defects generated in gamma-ray irradiated lithium oxide-

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In blanket systems for the D-T fusion reactor, it is considered that ternary lithium oxides will be introduced as a tritium breeder by reactions of

^{6}Li(n,α)T and ^{7}Li(n,n'α)T. After generation of tritium, it will lose its energy and be trapped by irradiation defects induced by high energy radiation. Many studies have reported that behaviors of irradiation defects during annealing processes are associated with tritium trapping and detrapping, and annihilation behaviors of irradiation defects associated with tritium release show two processes attributed by recombination or aggregation. Therefore, it is important to understand the annihilation behaviors of irradiation defects. On the other hand, gamma-ray irradiation can induce irradiation defects by only electron excitation process. Then, in this study, the annihilation kinetics of irradiation defects in gamma-ray irradiated Li_2O were clarified by Electron Spin Resonance (ESR). From the ESR spectra, F^+-center and O^--center were formed as irradiation defects. According to the isochronal annealing experiments, it was indicated that F^+-center and O^--center were disappeared by the recombination and F-center aggregation (FA) was formed at the temperature range of 300 - 475 K. The activation energy for recombination of F^+-center and O^--center was calculated by the results of isothermal annealing experiment to be 0.36 eV.

2B04: Mössbauer Spectra of New Cathode Active Material for Lithium Ion Battery

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Mössbauer spectra of cathode for lithium ion battery, in which $LiFeVPO_x$ glass is used as a cathode active material, show a reduction of Fe^{III} to Fe^{II} after discharge to 2.0 V, while an oxidation of Fe^{II} to Fe^{III} after charge to 4.0 V. Isomer shift of Fe^{III} (0.43 mm s^{-1}) and that of Fe^{II} (1.14 mm s^{-1}) indicate the presence of structural units of FeO_6 octahedra, as is the case for several phosphate glasses. Heat treatment of $LiFeVPO_x$ glass results in an increase in the specific capacity of the lithium ion battery from 50 to 150 mAh g^{-1}, due to a structural relaxation of the glass network accompanied by an increase in the number of polaron occupying the conduction band (CB). The author wishes the Mössbauer spectroscopy to be actively utilized for the resource and energy development.

2B05: Crystal structures and Mössbauer spectra of ZrO_2–$LnO_{1.5}$ (Ln = Dy, Yb) and CeO_2–$GdO_{1.5}$ solid solutions

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Powder X-ray diffraction (XRD) measurements of $Zr_{1-y}Ln_yO_{2-y/2}$ (Zr–Ln; Ln = Dy, Yb), prepared by a calcination at 1450 °C, show that Zr–Dy system forms a disordered defect-fluorite (F) phase ($0.12 \le y \le 0.65$) and C-phase ($0.80 < y$), while δ-phase ($Zr_3Yb_4O_{12}$) appears as mixed phases in the middle range ($0.42 \le y \le 0.80$), sandwiched with F-phase ($0.10 \le y \le 0.42$) and C-phase ($0.8 < y$). In Ce–Gd system, only single phases, F- ($y \le 0.30$) and C-phase ($0.30 < y$), are found. ^{155}Gd Mössbauer spectra of Ce-Gd system show that Gd^{3+} ions in C-phase occupy 24d site, oxygen defects (V_O) occuping face-diagonally in an octant, preferentially to the 8b site, V_O's at body-positions, and their occupancies increase rapidly with decreasing y from 1.0 to 0.50, and local structures become almost the same as those of F-phase below $y = 0.50$. The distortion of 8b site increases with increase in Ce content (from $y = 1.0$ to 0.50), while symmetry of 24d site increases gradually.

2B06: Observation of local fields in a perovskite $La_{0.7}Ca_{0.3}MnO_3$

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Perovskite manganese oxides are known to exhibit the effect of colossal magnetoresistance (CMR), a phenomenon that electrical resistivity undergoes a drastic change as large as five to six orders of magnitude by the magnetic transition. Because of this unique physical property, much attention has been given to these oxides aiming at wide industrial applications. In order to realize practical use of these compounds, it is of importance to obtain information on local fields in the material as well as macroscopic quantities such as resistivity and magnetization. From this point of view, we have applied the time-differential perturbed angular correlation (TDPAC) method to a study of a perovskite $La_{0.7}Ca_{0.3}MnO_3$ (T_c~245 K). Powder

$La_{0.7}Ca_{0.3}MnO_3$ was synthesized by a conventional solid-state reaction in air. For the introduction of the parent nuclides of the ^{111}Cd probe into the sample, two different ways were adopted: an ion-implantation method and a chemical synthesis. After the introduction of the parent nuclides, the sample was annealed at 1473 K and TDPAC measurements were performed. The TDPAC spectra suggest that the majority of the probe atoms reside in a unique site. In the presentation, the transferred field from Mn ions at the probe nucleus is discussed based on the temperature dependence of the spectra.

2B07: Activity ratio of ^{210}Pb/^{226}Ra in the volcanic products

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Observations were carried out on the activity ratio of ^{210}Pb/^{226}Ra in the products from Unzen (1993 AD eruption), Sakura-jima (1779 AD, 1946 AD, 1976 AD, 1977 AD and 1978 AD eruptions) and Izu-Oshima (1421 AD eruption) volcanoes, Japan. Measurement of ^{210}Pb/^{226}Ra activity ratio in the samples was made by gamma-ray spectrometry. The ^{210}Pb/^{226}Ra activity ratio in the samples ranged from 0.9 to 3.7. It is suggested that the ^{210}Pb/^{226}Ra < 1 (^{210}Pb deficits) in the samples result from the continuous degassing of ^{222}Rn, a precursor of ^{210}Pb in the decay series, from the shallow magma chamber. On the other hand, the ^{210}Pb/^{226}Ra > 1 (^{210}Pb excesses) in the samples are often attribute to the accumulation of ^{222}Rn in the magma chamber. The samples with large ^{210}Pb excesses, however, have an observable amount of ^{137}Cs, which indicates the contamination of the deposition of atmospheric ^{210}Pb.

2B08: Determination of the growth rate of precious corals using the natural radionuclide

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Lead-210 is a naturally occurring radionuclide of the U-238 decays series with a half-life of 22.3 years. ^{210}Pb is a natural marker suitable for dating events that have occurred over the past 100 years and has been actually used for measuring sedimentation rates of lake and coastal sediments. The objectives of this study are to measure the vertical distributions of ^{210}Pb concentrations in precious corals and to estimate the radial growth rate. The radial growth rate of the skeleton can be estimated by the gradual decreases in ^{210}Pb concentrations measured from the surface inwards. The radial growth rate of the red coral skeleton is estimated to be 0.22 mm/year, so slow that it would take as long as 50 years for a colony to grow to 22 mm in diameter.

2B09: Nuclear reaction rates for nucleosynthesis of the p-nucleus Sm-146

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The p-nuclei are not able to be produced from neutron capture process because they are neutron deficient. They are supposed to be produced in the network reaction including emission of neutron or capture of proton above temperature of 1×10^9 K. We focus on the Sm-146 isotope, which was abundant in the early history of the solar system, from view point of the astrophysical nucleosynthesis. There are some possible paths to synthesize the isotope through ^{147}Sm(γ,n), ^{147}Sm(γ,pβ$^-$), and ^{146}Nd(p,nβ$^-$), etc. We have measured cross sections of ^{147}Sm(γ,n)^{146}Sm and ^{147}Sm(γ,p)^{146}Pm and ^{146}Nd(p,n)^{146}Pm to investigate the paths and the ^{146}Sm production rate of each path.

The isotopes of ^{146}Sm and ^{146}Pm produced in bremsstrahlung irradiation and proton irradiation were measured by gamma spectrometry and accelerator mass spectrometry to obtain the cross sections.

As a results, we determined the cross sections of ^{146}Nd(p,n)^{146}Pm down to 3.5 MeV. The cross sections of this work are compared with theoretical calculation using the NON-SMOKER code for each incident energy and used to estimate astrophysical reaction rate. The data are useful to discuss the synthesis path to the ^{146}Sm isotope from comparison between astrophysical reaction rates of ^{146}Sm(γ,n)^{146}Sm, ^{146}Sm(γ,p)^{146}Pm, and

^{146}Nd(p,n)^{146}Pm.

2B10: Study on transport of dissolved organic matter in the Tokachi River system

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Humic substances are the largest fraction of dissolved organic matter in natural waters and consist of humic (HA) and fulvic acids (FA). The characteristics of aquatic HA and FA are important to understand the sources and dynamics of aquatic humic substances in river systems. This study reports the downward trend in Δ^{14}C and δ^{13}C of dissolved HA and FA isolated from river waters along the Tokachi River, which is located in Hokkaido Island in Japan. The HA and FA were isolated from the river waters at three stations from the upper to lower Tokachi River using the XAD extraction method in 2003-2005. Stable carbon isotopic ratio is determined by mass spectrometry and is reported in δ^{13}C notation versus the VPDB standard. Radiocarbon in humus samples was determined using accelerator mass spectrometry at the JAEA and NIES. The Δ^{14}C and δ^{13}C values of FA are younger and lighter than those of HA. The Δ^{14}C of fulvic acids decrease from +22‰ in the upper site to –58‰ in the lowest site. The downward trend is similar with that of HA. These results suggest that the supply of older HA and FA from watershed increase downward along the Tokachi River.

3B01: Radiocarbon age, calligraphical style and kiwamefuda certificate of kohitsugire calligraphy

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Radiocarbon ages of wooden samples generally disagree with their archaeological or historical ages mainly because the samples made from long-lived species or seasoned wood indicate old radiocarbon ages. Although Japanese paper is also wooden sample, it was made from branches of shrubs. Old paper is seldom used for calligraphy because it repels India ink and is unsuitable for elegant handwriting. Therefore, it can be considered that Japanese paper is a suitable sample for radiocarbon dating. In this study, we measured radiocarbon ages of ancient Japanese calligraphies called kohitsugire and documents of known age. The results demonstrated that the radiocarbon ages correspond to the years in which they were written. Most of kohitsugire have traditional certificates (called kiwamefuda) which identify the calligraphists. Paleographical study, however, shows that there are some kohitsugire whose calligraphical styles are incompatible with the descriptions of the kiwamefuda certificates, for example, kohitsugire attributed to Nakatoni no Kamatari (614-669) by the kiwamefuda seems to be written in the 10th – 14th century, and kohitsugire attributed to Prince Munetaka (1242–1274) has a calligraphical style of the 11th century. Therefore, in this study, we applied radiocarbon dating to these kohitsugire. Their radiocarbon ages corresponded to the paleographical ages and suggested that kiwamefuda does not necessarily agree with the years in which the calligraphy was written.

3B02: Measurements of ^{36}Cl/Cl in soil by using multi-step extractions

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Chlorine in soil is present in various chemical forms. From our previous studies, it has been confirmed that depth profiles of ^{36}Cl/Cl isotopic ratios for chlorides which have higher fluidity underground show maximum around surface layer. In this work, we tried to investigate the relationship between several types of chlorine which was supplied from different reservoirs. The following extraction methods were carried out with surface soils of several sites and ^{36}Cl/Cl isotopic ratio of each sample was measured with accelerator mass spectroscopy (AMS) at UTTAC;

A) Chlorides were extracted with dilute nitric acid (inorganic soluble phase).

B) Organochlorines were resolved with hydrogen peroxide after acid extraction (organic phase).

C) Chlorine in minerals was taken out by combustion from residue (mineral phase).

It was found that some sort of interaction between chloride and organochlorine in soil was considered from the ^{36}Cl/Cl ratio. On the other hand, the ^{36}Cl/Cl ratio in mineral phase showed features different from soluble and organic phase. There might be another process on existing in mineral phase which is different from soluble and organic phase.

3B03: Studies on the secular variation of ^{129}I/^{127}I ratios in the atmospheric fallout in Tokyo and Akita using AMS

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Iodine-129 is one of the most important radionuclides in environmental safety. In this study, we have developed an appropriate separation procedure of iodine from atmospheric fallout samples for determining ^{129}I/^{127}I ratios by accelerator mass spectrometry (AMS). Samples used in this study were collected from Tokyo and Akita from 1963 to 2003 by Meteorological Research Institute.

The analytical results showed that the secular variation of ^{129}I/^{127}I ratios in the atmospheric fallout was different from that of ^{90}Sr and ^{137}Cs. This suggested that the variation of ^{129}I/^{127}I ratios observed was not due to the nuclear bomb tests. High ^{129}I/^{127}I ratios (up to 8×10^{-8}) found after 1980 indicate that contribution from the other sources (e.g. possibly from nuclear spent fuel reprocessing plants) might be important to understand the increase of the ratios. Moreover, an increasing tendency of ^{127}I in the atmospheric fallout with time was observed, of which reason is still unknown.

3B04: Determination of iodine in estuarine biota by ICP-OES

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Some marine biota are known as iodine accumulators, and thus they could also absorb a long-lived radioiodine (iodine-129) released to the marine environment. However, not all marine biota contain high amount of iodine. In this study, estuarine biota samples were used to develop a simple iodine measurement method. A 0.1-g portion of each sample was weighed in a PFA vial, and Milli-Q water and Tetramethylammonium hydroxide (TMAH) were added. The mixture was heated at 60 ˚C for 15h and then diluted with Milli-Q water. Finally, iodine in the solution was measured by inductively coupled plasma optical emission spectroscopy (ICP-OES).

There are three major wavelengths for iodine; however nitrogen and phosphorus have neighboring lines to two wavelengths of iodine. Therefore 182.976 nm was selected for an optimum wavelength for iodine analysis. The detection limit of the present method was 150 mg/kg when a 0.1-g of sample powder was used. The results for a standard reference material (NIES No. 9, reference value: 520 mg/kg) obtained by this method showed a good agreement (544±44 mg/kg). This method could be successfully applied to determination of iodine in estuarine biota.

3B05: Application of ^{85}Kr dating to groundwater in Kumamoto Area, Japan

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Groundwater age was determined by ^{85}Kr/Kr specific activity of gases dissolved in groundwater at Kumamoto Area, in which newly developed Kr extraction system and liquid scintillation counting technique for ^{85}Kr were applied. Apparent mean residence times observed were 8.2 ± 0.7 y and 20.7 ± 0.6 y for the groundwater taken from a well at recharge area and that from an artesian borehole at the discharge area, respectively showing a consistent age trend estimated from the observed groundwater flow system of Kumamoto Area and also that determined using environmental tritium. Similar mean residence time of ^{85}Kr and tritium was observed at the discharge area but the mean residence time of ^{85}Kr at the recharge area was younger compared to that of tritium, probably due to mixing of river water containing recent high ^{85}Kr concentration in this region into groundwater flow coming from the northern upland's recharge area. The mean residence time of the groundwater flow coming from northern upland area and a traveling time of the groundwater between two sampling points were estimated by a simple model assuming mixing the river water with the groundwater and a piston flow between two sampling points.

3B06: An approach toward identification of radioxenon sources for nuclear explosion verification

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Radioxenon nuclides including ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe have been used for detection of nuclear explosion, because these nuclides can be easily leaked out from even underground due to their large fission yield and inert chemical property. Twenty seven radioxenon monitoring stations have been installed in the world including Takasaki, Japan, to detect the nuclear explosion as a verification system of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). Irrelevant radioxenon to the nuclear explosion has been occasionally detected at the monitoring stations, which makes difficult to distinguish from the nuclear explosion. These are considered as the radioxenon sources: nuclear power and research reactors; spent fuel reprocessing facilities; nuclear particle accelerators; hospitals; natural background from lithosphere; and flow from oversea. We estimate annual discharged radioxenon activities of each emission source, except for the flow from oversea, using published information and evaluate the radioxenon counts of the Takasaki monitoring station. An approach toward identification of radioxenon sources using atmospheric transportation modeling and radioxenon emission data is also presented.

3B07: Separation of Ba and Ra using the difference of their adsorption to manganese dioxide

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In general, the quantitative determination of radium in water sample is carried out by gamma-ray measurements of precipitate obtained by coprecipitation of radium with barium sulfate. Barium reagents usually contain some amount of radium. Therefore the use of barium reagent is limited due to the interference of radium in the barium reagent. In this work, we have investigated the separation of radium from barium reagent using the difference of adsorption properties of barium and radium cations to manganese dioxide. The adsorption rate and the distribution ratio for the elements show that manganese dioxide adsorbs radium cations in preference to barium cations and that the adsorption of barium and radium cations follows the stoichiometric ion-exchange mechanism.

3B08: Plating method of Po from acid solutions onto a stainless-steel disk

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A plating method has been devised for trace amounts of polonium (Po) from acid solutions with high yield onto a stainless-steel disk. The conditions for effective deposit of Po were examined for an electrodeposition method other than the spontaneous deposition method. In this work, ^{209}Po isotope was added to electrolysis solutions, and Po recovery was measured under various conditions such as plating time, electrolysis current, temperature, major anion species, and co-presence of other metal-ions. It was found that the electrodeposition method provided a good recovery and selectivity from HCl (0.5-6 M), HNO_3 (0.5-2 M), H_2SO_4 (0.5-6 M) and H_3PO_4 (0.5-6 M) solutions. The optimized conditions for Po deposition were 0.2-0.3A (~100 mA/cm^2) in electrolysis current, plating for 2 hours in room temperature. Besides, interfering ions were not deposited in acid solutions over 0.5 M. The electrodeposition of Po from acid solution is concluded to be more effective and rapid, than the spontaneous deposition.

3B09: Optimization of Automated Sequential Anion-Exchange Separation of Ultra-trace Elements

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From these ratios of trace U, Th, Pb and lanthanides in an environmental sample, information on origin and history of the sample can be obtained. The key elements should be separated from the co-existing and interfering elements in a sample to analyze accurately the abundance and isotopic ratios of the key elements by ICP-MS. The sequential separation technique of these elements with a single anion-exchange column has been developed. In this work, the pressurized sequential separation system with compressed air, a switching valve, and Teflon tubes was examined to make up an automated separation system. An eluent in a Teflon tube was introduced to an anion-exchange column with pressurized air. The eluents were changed by a switching valve. The separation was optimized for some parameters including the flow rate of eluents and the composition of eluents. The mixture of CH_3COOH and HNO_3 was effective for the separation of Fe from U. The composition of 65 vol.% CH_3COOH and 1M HNO_3 was the best for the separation. This pressurized separation system improved the resolution of separation peaks and reduced the separation time compared with the previous gravity flow separation.

3B10: Development of elemental and isotopic analyses of uranium, thorium, and macro elements in the environment using an automatic SPE injection equipment coupled to ICP-DRC-MS

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Beginning in FY2009, funding for the equipment cost of ICP-MS analytical system was allowed to insert it in the budget of The University of Tokushima. The aim of equipping this new system is to educate the undergraduate students in the Department of Radiologic Science to understand and train advanced elemental and isotopic analyses, which are also available for their graduation researches on radiologic sciences. In addition, this system also will be effectively shared among biomedical researchers who are with faculties of medicine, dentistry, and pharmaceutical sciences at Kuramoto Campus in The University of Tokushima. In particular, the present ICP-MS analysis will be expected to contribute to measure quantitatively trace and macro amounts of metal elements (metallomics) relative to environmental radioactivity, biology, medicine, and so on. Until now, we have developed the integrated analysis system carried out from decomposing various samples, to isolate target elements automatically by a SPE (Solid Phase Extraction) method, and to measure their signal intensities quantitatively mass-separated using an unique ICP-DRC-MS (Dynamic Reaction Cell) type. In this presentation, we have reported the experimental condition under this development.

3B11: Feasibility of using globalfallout ^{236}U to construct the circulation of seawater

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The global fallout ^{236}U ($T_{1/2}=2.342\times10^{7}$ y, α-decay) level in soil was deduced from measurements of ^{236}U, $^{239+240}$Pu and ^{137}Cs in surface soils which are solely influenced by global fallout. A total of 12 soil cores from the depths of 0-10, 0-20 and 0-30 cm were collected at a flat forest area in Japan. Concentrations of $^{239+240}$Pu and ^{238}U were determined by α-particle spectrometry, while the ^{236}U/^{238}U ratio was measured with accelerator mass spectrometry (AMS). Consistent ^{236}U/^{239}Pu ratios between 0.212 to 0.253 were found. Using this ratio, the total global fallout of ^{236}U on the earth is estimated to be as much as ca. 900 kg. This knowledge will contribute to the promotion of research on U isotopes, including ^{236}U, for the fields of geo-resources, waste management and geochemistry. The application of global fallout ^{236}U to the water circulation in the "miniature ocean", Japan sea, will be discussed in our presentation.

3B12: Uptake of Uranium by Spinach Plants after Application of Elevated Amount of Phosphatic Fertilizers to Upland Field Soil

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Phosphatic fertilizers made from phosphate rocks usually contain high amounts of uranium so that it is sometimes listed as naturally occurring radioactive materials (NORM). It would be possible that plant can absorb U from this type of fertilizer just after its application to agricultural soils because U in the fertilizer may be more mobile than that originated from soil. In order to clarify the effect of phosphatic fertilizer application for U uptake by agricultural crops from soil, we cultivated spinach plants on six different fertilizer concentration fields in Okayama city. Concentrations of U in the plant at harvest, and those in soil samples for several times during planting period were measured. We found that concentrations of U in spinach samples and those of the extract from the soil with water were well correlated. Also, concentrations of U and Al in spinach were highly correlated. From these results, it was implied that spinach plants would absorb water soluble U with water through roots.

3B13: Thorium deposition observed at Tsukuba

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Monthly depositions of thorium isotopes at Tsukuba, Japan during the period 1990-2007 were determined. The monthly ^{232}Th deposition showed a typical seasonal variation with inter-annual variability. Especially, a marked peak of the ^{232}Th deposition as did ^{239,240}Pu occurred in spring. The seasonal change of the monthly ^{232}Th deposition is closely related to those of the Kosa events. High monthly depositions of thorium in spring might originate from soil particles, as did ^{239,240}Pu. However, the inter-annual variation of the annual ^{232}Th deposition did not coincide with that of the Kosa event frequency. The ^{230}Th/^{232}Th activity ratios in deposition samples showed large variability; its higher ratios were observed in early spring. The higher ^{230}Th/^{232}Th ratios reflect local soils contaminated by uranium originating from fertilizers, whereas the low ratios occur in lithogenic materials. Therefore, the ^{230}Th/^{232}Th 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. The local ^{232}Th deposition showed a smaller peak in early spring, corresponding to local dust storm event.