

English Abstracts

1S01 Basic Science and Applications of Nanocarbons for Green Innovation

Endo, M. (Faculty of Engineering, Shinshu Univ.)

Carbon nanotubes have attracted lots of attention from various fields of scientists because they exhibited extraordinary physical and chemical properties due to their intrinsic nano-sized one-dimensional nature. It should be noted that lots of carbon nanotube-derived products are already in use and their viability strongly depend on their commercialization. Among them, we have developed a super rubber sealant that is able to withstand temperatures up to 260°C and pressures as high as 239MPa by incorporating surface-modified carbon nanotubes into fluorine rubber. Thus our rubber sealant will contribute to a revolutionary enhancement in the oil recovery efficiency from the current 35 % to more than 70 % by excavating previously inaccessible deposits. However, the biggest hurdles in nanotube business are considered to be the safety issue of carbon nanotubes. By sharing the all information on risks and benefits of the materials with all stakeholders, we are able to prove the carbon nanotubes to be the green and safe innovative materials by responsible production and use in the 21st century.

2S04 Study on the environmental uranium isotope

Aya Sakaguchi (Hiroshima Univ.)

The uranium normally found in nature consists of three isotopes of mass numbers 234, 235 and 238. They exist ubiquitously on earth, and show the specific concentration and composition of isotope. By using these features, uranium isotope in the environment has been used as a fingerprint of the uranium contamination and tool for the study of geochemistry/geosciences.

Recently, the uranium isotope ^{236}U ($T_{1/2}=2.342\times 10^7$ y, α -decay) has been recognized as one of the “environmental Uranium(U) isotopes” due to the development of instruments and measuring techniques of sufficient abundance sensitivity, and applications which use ^{236}U as a proxy/tool for environmental and geochemical uranium studies are developing.

One of the main source of ^{236}U at the surface environment is global fallout which was introduced from the atmospheric nuclear test in 1960s. The global fallout ^{236}U level in soil was deduced from measurements of ^{236}U , $^{239+240}\text{Pu}$ and ^{137}Cs in surface soils which are solely influenced by global fallout. Soil cores of 30 cm depth were collected from undisturbed flat areas in Japan. Concentrations of $^{239+240}\text{Pu}$ and ^{238}U , atom ratios of $^{239}\text{Pu}/^{240}\text{Pu}$ and $^{236}\text{U}/^{238}\text{U}$ were determined by α -particle spectrometry, ICP-MS and accelerator mass spectrometry (AMS), respectively. Consistent $^{236}\text{U}/^{239}\text{Pu}$ 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.

2S01 Fukushima Daiichi NPP Accident, details of accident event and current status

YAMANA, H. (Research Reactor Institute, Kyoto University)

Due to the massive earthquake and devastating tsunami happened on March 11th, 2011 Fukushima Daiichi

Nuclear Power Plant suffered serious loss of core cooling function. It led to extensive core meltdowns of three units, and to serious building failures caused by hydrogen explosion which was generated from the severe accident process. As a result, massive amount of radioactive nuclides were released to the atmosphere resulting in the large regional contamination and radiation exposure to the public. The restoration of the plants is ongoing, and the accomplishment of cold shut down of three units is expected before the end of January 2012. Discussions on the possible measures to be taken after the completion of current restoration program, has been started under the frame of Atomic Energy Commission. It will draw a roadmap for further restoration and clean-up, including removal of spent fuels, inner inspection of the damaged cores, as well as removal of damaged fuels and debris.

This paper reviews the accident of Fukushima Daiichi Nuclear Power Plant, in terms of the technical details of accident process, current status, and future prospect of restoration.

2S02 Behavior of radionuclides in the environment and their transfer to foodstuffs.

Y. Muramatsu (Gakushuin Univ.)

The most important radionuclides released from the accident of Fukushima Daiichi are Xe-133 (half-life: 5.2d), I-131 (8.0d), Cs-134 (2.1y) and Cs-137 (30y). Although the amount of Xe-133 released was very large, this nuclide is not so important related to radiation safety, because of its non-reactivity to vegetations and soils. I-131 is important in the first one month and high concentrations in leafy vegetables were observed. However, due to its short half-life, the concentration levels in agricultural crops decreased with time. In case of radiocesium, attention should be paid to the uptake of this nuclide from soil to plants. At the beginning of April, the government recommended a guideline for the maximum concentration of radiocesium in soil as 5000 Bq/kg. This value was based on the soil-rice transfer factor of 0.1, which was estimated from the values of previous studies in Japan. Radiocesium concentrations in some agricultural crops (tea, apricot, mushrooms, etc) exceeded the guideline for radiocesium in foodstuffs (500 Bq/kg). Mechanisms in the transfer of the nuclide in to these crops have been studied.

I will summarize behavior of radiocesium and some other nuclides in the environment and possible pathways to foodstuffs and drinking water.

2S03 Biological effect of radiation: basis for understanding the risk of Fukushima Nuclear Accident

Imaoka, T. (Nat. Inst. Radiol. Sci.)

The radionuclide release in the Fukushima Nuclear Accident has induced a tremendous anxiety on possible health effects of low dose radiation. When radiation hits a cell in an organism, it may induce DNA damages which, if not repaired properly, lead to either cell death or genetic mutation. If function of the tissue is lost as a result of cell death, various tissue responses including dysfunction of hematopoietic tissues, sterility and skin responses may occur; these responses are not manifested if the radiation dose is low enough. Genetic mutation is considered to occur, albeit at a low frequency, even if the radiation dose is very small. Cancer is a result of genetic mutation and its

probability is considered to rise, albeit slightly, if radiation induces a small amount of additional mutations. These assumptions lead to a notion that there is no “safety dose” below which radiation does not cause any cancer. On the other hand, the study of atomic bomb survivors of Hiroshima and Nagasaki provides the most reliable quantitative information on the relationship between radiation dose and accompanying increase in cancer risk. The analysis so far indicates that cancer risk increases by 0.5-fold, compared to a background level, if a human body is exposed to 1 sievert of radiation; at lower doses, the risk is proportional to the dose, but it is impossible to detect cancer risk associated with 100 millisievert of exposure because of statistical limitations. Although exposure to atomic bomb radiation occurred in a very little instance, the current situation poses a prolonged (i.e., low dose rate) exposure, probably resulting in still lower cancer risk. Still, since current radiation exposure has no benefit, unlike that in medical situations, it is important to reduce it to a level as low as reasonably achievable.

I will explain the biological effect of radiation, including its mechanistic basis and effects on the human body, and wish to help the audience to understand the impact of the accident on public health.

1T01 Characteristics of distribution of radionuclides from Fukushima Daiichi Nuclear Power Station in Fukushima prefecture and neighbor prefectures obtained by a gamma-ray measurement immediately after the accident

Matsumura, H., Saito, K. (KEK), Ishioka, J., Uwamino, Y. (RIKEN)

In order to investigate diffusion of radioactive materials from the Fukushima Daiichi Nuclear Power Station, in Fukushima and neighboring prefectures, the dose rates and γ -ray spectra were measured on expressways on March 15, 16, 17, and April 8, 2011. A 1.5-inch LaBr₃ γ -ray spectrometer was used for the measurement. A radioactive plume was observed at Koriyama-higashi interchange on the afternoon of March 15. The radioactive materials arrived in the Nakadori region of Fukushima prefecture was deposited by rainfall on the afternoon of March 15. Although the distributions of ¹³²Te, ¹³²I, ¹³⁴Cs, ¹³⁶Cs, and ¹³⁷Cs were similar, the distribution of ¹³¹I was different from the others. There were low ¹³¹I deposition areas in the Nakadori region of Fukushima prefecture.

1T02 Survey of the radiation by a car borne survey method and γ ray spectrometry by NaI detector in Fukushima Prefecture

Sugihara, S., Ichikawa, R., Maekawa, A., Matsuzaki, T., Kawamura, H., Momoshima, N. (Kyushu Univ.)

The Tohoku District-off the Pacific Ocean Earthquake and Tsunami caused by the earthquake attacked the Fukushima Dai-ichi Nuclear Power Plant of TEPCO on March 11, 2011. A nuclear accident followed at an unprecedented scale and huge amounts of radioactive material were released into the environment.

Distributions of dose rate in Fukushima prefecture were measured on April 18-21 and June 20-22, 2011 by a car borne survey method using a NaI(Tl) scintillation survey meter. The gamma-ray spectrometry by a NaI(Tl) detector was also done at several points. Dose rate near Fukushima

Dai-ichi NPP and at Iitate-mura, Fukushima-city was high (1 ~ >30 μ Sv/h). So distribution of dose rate is uneven, further investigation is necessary. The gamma-ray peak of I-131, Cs-134 and Cs-137 were mainly detected in gamma-ray spectrum.

1T03 Radioactivities from the accident of Fukushima 1 Nuclear Power Plant - Observation at Radioisotope Center, University of Tsukuba.

Sueki, K., Suzuki, M., Sakamoto, K., Ito, T., Furukawa, J., Oshio, H., Matsumoto, H. (RIC, Univ. Tsukuba)

A tremendous amount of radioactivity in the atmosphere and in the sea was discharged due to the core meltdown accident of nuclear reactors in the Fukushima No.1 nuclear power plant in March 2011. The radioactive substance has reached Tsukuba away about 170 km in the south-southwest from the nuclear power plant. It reported that the effective dose rate measured at the radiation monitor and the radioactivities measured the collection of atmospheric suspended dust used by low volume air sampler, the collection of rain water, and surface soil at Radioisotope Center, Univ. of Tsukuba. The analysis of γ -ray spectroscopy confirmed the radionuclides such as ¹³¹I, ¹³²Te (\rightarrow ¹³²I), ¹³⁴Cs, ¹³⁶Cs, and ¹³⁷Cs. We report the time variation of the radioactivity concentrations of several radionuclides at Univ. of Tsukuba.

1T04 Radioactivity in atmospheric particulates collected at Hachioji, Hino, and Kazo.

Oura, Y., Matsuda, K., Yonemochi, S., Igarashi, Y., Ebihara, M. (Tokyo Metropolitan Univ., Meisei Univ., Center for Environ. Sci. in Saitama, Meteorological Research Insti.)

The radioactivity in PM_{2.5} atmospheric particulate matters collected at Hachioji, Hino, and Kazo cities after Great East Japan Earthquake have been measured. At Hachioji, total suspended particulate matters (TSP) and gaseous matters were also collected. Radioactivity in atmospheric particulates decreased with high concentration peaks on 15 Mar, 21-22 Mar., 30 Mar., and 17 Apr. Radionuclides leaked from crippled nuclear power plants were found to be concentrated in fine particulates comparing PM2.5 with TSP and ¹³¹I suspended as gaseous species in the air were about 4.5 times in average more abundant than as solid species. It is simply estimated that citizen in Hachioji intake about 35 kBq of ¹³¹I and 5.4 kBq of ¹³⁴⁺¹³⁷Cs after the nuclear power plant accident.

1T05 Atmospheric radionuclides detected at Fukuoka, Japan released from the Fukushima Daiichi nuclear power complex following the nuclear accident

Momoshima, N., Sugihara, S., Ichikawa, R., Yokoyama, H. (Kyushu Univ.)

Radionuclides from the Fukushima nuclear accident were detected at Fukuoka, Japan, 1000 km distance westerly from the Fukushima Daiichi nuclear power complex. The first arrival of ¹³¹I was confirmed on March 17, 2011 within 3 days after the release, indicating the ¹³¹I was probably transported to Fukuoka dispersively due to local meteorological condition, not a global air circulation. A maximum concentration, as much as 5.07 mBq m⁻³ for ¹³¹I, 4.04 mBq m⁻³ for ¹³⁴Cs, 4.12 mBq m⁻³ for ¹³⁷Cs was recorded in particles collected on April 6, 2011; however the level decreased below the detection limit after April 23,

2011. Gaseous ^{131}I occupied 30 % to 67 % of the total ^{131}I . The increase in internal dose by inhalation was negligible at Fukuoka.

1T06 Radiochemical Analysis of Fission and Neutron Activation Products released by Fukushima Nuclear Power Plant Accident.

KOJIMA, S., ARINOBU, T., KOSUDA, C., KATO, M. (Aichi Med. Univ.), FURUKAWA, M. (CNIC)

This paper describes the determination of the composition and content of fission and neutron activation products in the environment released by Fukushima Daiichi Nuclear Power Plant Accident. A soil sample was collected on May 19 at Namie-cho Sakai, Fukushima Prefecture, 8 km northwest from Fukushima Daiichi Nuclear Power Plant. Non-destructive gamma-ray spectrometry had been performed with a well-type Ge detector before radiochemical analysis, and six gamma-ray emitting radionuclides, ^{137}Cs , ^{134}Cs , $^{129\text{m}}\text{Te}$, $^{110\text{m}}\text{Ag}$, ^{136}Cs and ^{140}Ba (^{140}La) were measured in the soil sample. The high radioactive concentrations of ^{137}Cs and ^{134}Cs were obtained to be 1880 and 1530 $\text{Bq}\cdot\text{g}^{-1}$, respectively. Radiochemical separation by precipitation, solvent extraction and anion-exchange removed those radionuclides. Analytical results indicate that the radioactive contents of fission products, ^{141}Ce , ^{144}Ce and ^{125}Sb were ranged from four ten-thousandths to one thousandth relative to the amount of ^{137}Cs . Four neutron induced radionuclides, $^{114\text{m}}\text{In}$, ^{54}Mn , ^{58}Co and ^{60}Co , were detected. The concentration of $^{114\text{m}}\text{In}$ was about one thousandth to ^{137}Cs , and the others were less than one tenth relative to $^{114\text{m}}\text{In}$. We conclude that $^{110\text{m}}\text{Ag}$ and $^{114\text{m}}\text{In}$ were produced by the neutron capture reactions in nuclear control rods made of silver-indium-cadmium alloys.

1T07 Spatial distribution of Iodine-129 in surface soil around the Fukushima Daiichi nuclear power plant

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Due to the accident at the Fukushima Daiichi nuclear power plant, which was caused by the Great East Japan Earthquake, a lot of radioactive materials were released into the environment. Among them, Iodine-131, which has a short half-life of 8 days, is thought to be hardly detected after the accident is concluded. It is very important to research how leaked out Iodine-131 was diffused in order to estimate the health impact of radiation at the time of the accident. On the other hand, Iodine-129, which was leaked out and was thought to act chemically-identically as Iodine-131, has an extremely long half-life of 15.7 million years and we are able to measure it equally after the accident. By following the trail of Iodine-129, it is considered to estimate the distribution of Iodine-131. To do this, at first, it is essential to measure simultaneously Iodine-131 and Iodine-129 in the same sample picked from near-the Fukushima Daiichi nuclear power plant and examine the relation between them (for example, the isotopic ratio of Iodine derived from the nuclear power plant (I-129/I-131)).

At this study, we measured Iodine-129 in surface soil within 60 kilometers of the Fukushima Daiichi nuclear power plant, which was picked by research team of Nuclear

Engineering Research Laboratory, Faculty of Engineering, The University of Tokyo. We discuss Iodine-129 derived from the nuclear power plant by considering the concentration range, the relation of a distance or a direction from the nuclear power plant, and the relation between I-129 and other radioactive nuclides (Cs-134 , Cs-137 , I-131). Since Iodine-129, which had been leaked out from the nuclear fuel reprocessing plant in Europe, was already transferred to Japan by way of the atmospheric transportation before the accident, it is important to distinguish between Iodine-129 from this accident and from the reprocessing plant. Then, we want to obtain the I-129/I-131 ratio originating in the accident precisely and discuss the relation between the fuel burn-up and it.

As the way of experiment, iodine was extracted from soil and mixed with a carrier and I-129/I-127 ratio was measured by means of AMS (Accelerator Mass Spectrometry) at MALT (Micro Analysis Laboratory, Tandem accelerator, The University of Tokyo). Iodine-131 was measured by a germanium detector and Iodine-127 was measured by ICP-MS.

As a consequence, there appeared a certain amount of correlation between I-129 and I-131 from soil. However, about the isotopic ratio, there seemed variability. In this study, we discussed, as factors of variability, the incomplete homogeneity of soil samples treated, the yield of recovered Iodine when we burned soil, and types of soil. Because analysis samples are limited at this time, we don't still reach the conclusion. But, we obtained some knowledge and we intend to report them.

1T08 Radioactivity survey on the Fukushima nuclear accident in Fukushima and Western Kanto

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A tremendous amount of radioactivity was discharged due to the damage to cooling systems of nuclear reactors in the Fukushima No.1 nuclear power plant in March 2011. Fukushima and its adjacent prefectures were contaminated with fission products from the accident. We'll show geographical distributions of individual radionuclides in the surface soils of central-east Japan as determined by gamma ray spectrometry. Especially in Fukushima prefecture, contaminated area spreads around Iitate, Namie, and Naka-Dori for the all radionuclide we measured. Distributions of the radionuclides were affected by the physical state of each nuclide as well as geographical features. Considering meteorological conditions, it is concluded that the radioactive material transported on 15 March was the major contributor to contamination in Fukushima prefecture, while that transported on 21 March was the major source in Ibaraki, Tochigi, Saitama, and Chiba prefectures.

1T09 Studies on the transfer of radiocesium into fruits during their growth

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T.Ohtsuki² (¹Gakushuin Univ., ²Fukushima Agri. Tech. Ctr., ³Tohoku University)

Concentrations of radiocesium in several fruits collected from Fukushima-Prefecture and other areas in Japan were determined with Ge-detectors. It was found that Japanese apricot (Ume) had relatively higher radiocesium concentrations compared to other fruits. This could be explained by the fact that they were blooming at the major release of the nuclides and radiocesium was deposited in the blossoms, which resulted in the contamination. In case of other fruits such as peaches, cherries, grapes etc, some enhancements of the concentrations were thought to be due to the translocation from the barks, which were contaminated with radiocesium. The concentrations of radiocesium in the fruits such as peaches and cherries decreased with their growth. Mechanisms of radiocesium contamination of fruits and also some other plants (e.g. bamboos and tea) will be discussed.

1T10 Depth profiles of radioactive cesium and iodine released from the Fukushima Daiichi nuclear power plant and their transfer to the agricultural crops

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In order to understand the behavior of the radionuclide released from the Fukushima Daiichi nuclear power plant, we have measured depth distributions of radioactive cesium and iodine in different agricultural fields and forests. Our results demonstrate that more than 90% radionuclides were distributed from the surface to 6cm in depth. It is important to investigate transfer of the radionuclide to the agricultural crops. In this study, we have also determined transfer factors of radiocesium to agricultural crops such as cucumbers, tomatoes, eggplants and green peppers using the soils collected in Fukushima prefecture.

1T11 Depth profile of ¹²⁹I concentration in soil at Abukuma, Fukushima

Honda, M. (Nihon Univ.), Matsuzaki, H., Yamagata, T., Tuchiya, Y., Nakano, C. (The University of Tokyo), Matushi, Y. (Kyoto Univ.), Nagai, H. (Nihon Univ.).

Two 30cm forest soil core sample were collected before the Fukushima Daiichi nuclear power plant accident (ABK2) and after the accident (ABK3), and a field soil core sample (ABK12) was collected after the accident at Abukuma, Fukushima. The depth profile of ¹²⁹I concentration in soil at Abukuma, Fukushima have been measured by AMS at MALT, the University of Tokyo. At the top of 1.5cm, ¹²⁹I/¹²⁷I ratio in ABK3 was five times higher than ABK2, which imply accumulation of ¹²⁹I ejected by the Fukushima Daiichi nuclear power plant. For the other depth of these cores, ABK2 and ABK3, almost same depth profiles of ¹²⁹I/¹²⁷I were observed. The surface ¹²⁹I/¹²⁷I ratio in ABK3 was 31 times higher than Tamatsukuri, Miyagi (1.8×10^{-9} , Y. Muramatsu et al. 2007) and 1/5 of the Tokai-mura, Ibaraki (3.0×10^{-7} , Y. Muramatsu et al. 1985).

Since ABK12 is well mixed paddy field, depth profile of ¹²⁷I concentration was constant and ¹²⁹I concentration was lower than forest soil (ABK2, 3). ¹²⁹I/¹²⁷I decreases rapidly from surface to 9cm, and then became constant. From this

result, we considered accident origin ¹²⁹I were accumulated in the 9 cm of surface layer, and the ¹²⁹I inventory was estimated to 5.68×10^{11} atoms m⁻².

1T12 Removal of cesium from radioactive contaminated sea water using ferrocyanide-containing adsorbent

Suzuki, T., Kaneshiki, T., Nomura, M. (Tokyo Tech)

A large quantity of radioactive contaminated sea water was generated, because the sea water was used to cool the reactor emergently at Fukushima nuclear power plant accident. This radioactive contaminated sea water is preferable to be discharged into the ocean after the removal of the radioactive nuclides. In the present work, the removal of cesium from radioactive contaminated sea water was investigated using the ferrocyanide-containing adsorbent. Three types of ferrocyanide-containing adsorbents were tested. One is the adsorbent containing nickel ferrocyanide fixed by resin, which is provided by Purolite. Another is the adsorbent containing Prussian blue (ferric ferrocyanide) fixed by resin, which is manufactured by Nihon Kaisui Co., Ltd. Last one is the zeolite-supported ferric ferrocyanide, which is manufactured in our laboratory on trial. The adsorption properties of cesium on these adsorbents were estimated by batch experiment using sea water with 10ppm of stable cesium ion. It was confirmed that the either adsorbents have the large distribution coefficient and sufficient property of removal of cesium from sea water.

2T01 Radiation education in elementary and junior high school

Nakajo, S. (Grad. School of Edu., Shinshu Univ.)

Ministry of Education, Culture, Sports, Science and Technology (MEXT) revised the Courses of Study for elementary and junior high schools in March 2008. In this revision, the radiation education was introduced again after an interval of few decades. It is emphasized to make reference to the following points as part of learning about energy resources.

Nuclear power is the energy released from nuclear fuels, such as uranium.

Nuclear fuels emit radiation, but radiation is also present in nature.

Radiation has a transparency that is utilized in medical, manufacturing fields, and so on.

On the other hand, the Fukushima Daiichi Nuclear Power Station accident caused by the earthquake on March this year has made most people take a great deal of interest in radiation and radioactivity. A large majority of people wants to acquire proper knowledge and general understanding of radiation. Under these circumstances, several educational research works have been carried out in classes of elementary and junior high schools.

2T02

2T03 For education of radiation as science

Kudo, H. (Tohoku Radiation Science Center)

Disastrous release of radionuclides from Fukushima Daiichi Nuclear Power Station in the aftermath of big earthquakes and tsunamis on March 11, 2011 is giving hard influence on Japanese daily lives. People's attention is focused on health concerns, but most people are confused

about what they should do, because of a lot of inadequate information given by a variety of mass media that exaggerate the risk of radiation. At present, therefore, adequate social and school education of radiation based on science is required. Scientific knowledge of radiation and its reactions with matter would help people decide their rational behavior, not to be in a panic. Educators should keep in mind the adequate use of terminology in the course of education; e.g., discrimination of *radiation* and *radioactivity*. Good news is resumption of a radiation class in the science curriculum of middle high school in the next year after 3 decades of suspension.

2T04

2T05 Societies' Response to the nuclear power plant accident in Fukushima – Cooperation is essential

Ebihara, M (Grad. School of Sci. & Eng., Tokyo Metropolitan Univ.)

A gigantic earthquake (M 9.0) and resulting tsunami hit the Japanese islands on March 11, 2011. Besides direct disaster caused by earthquake and tsunami, equally serious accident happened at Fukushima Daiichi nuclear power plant of Tokyo Electric Power Company. By this accident, nuclear power reactors couldn't been controlled properly and eventually radioactive materials were released from the building to house nuclear power reactors and dispersed over rather wide area. Consequently, agricultural materials, drinking water and food were contaminated with radioactive nuclides. It is urgent to figure out how radioactive materials were spread not only in the area close to the nuclear reactors but also over surrounding, rather wide district. Such information must release the uneasiness for public people and objectively predict the possible influence caused by radioactive material over agricultural products for people engaging in agriculture.

To grapple with this task promptly as well as effectively, scientists belonging to the Geochemical Society of Japan, the Japan Society for Nuclear and Radiochemical Sciences and the atmospheric and oceanic section of the Japan Geoscience Union were allied and presented a proposal to MEXT for comprehensive and systematic survey of radioactive materials in the environment. In this project, samples are to be collected for atmosphere, rain, soils and underground water as systematically as possible and as wide as possible for the sampling area. As for soil samples, we decided to work together with volunteer people of nuclear physics, who also took similar actions to ours. After sampling, samples were subjected to precise measurement of radioactivity under controlled counting protocol and data thus obtained are to be opened to the governmental sector as well as public promptly. At the same time, scientific meaning and information derived from such data are to be reported openly to appropriate academic societies as well as to the public. From this experience, we acknowledged that cooperation of related societies is essential for recovering such chaotic circumstances as those happened in east Japan this year.

1P01 Correlation between Asian Dust (yellow dust) and specific radioactivities of fission products including in aerosol in Tokushima, Shikoku Island, due to the Fukushima nuclear accident

Sakama, M. (School Health Sci., Univ. Tokushima), Saze, T. (RIC, Univ. Tokushima), Sakaguchi, Y., Fushimi, K., Nakayama, S. (Institute Socio Arts and Sci., Univ. Tokushima)

The serious accident of the Fukushima Dai-ichi nuclear power plant in the southeast region of Tohoku, eastern Japan, was caused due to great tsunami following by the East Japan Earthquake on March 11, 2011, at around 14:46 JST. The airborne radioactive isotopes from the Fukushima Dai-ichi nuclear power plant were measured by analyzing aerosols at Tokushima, Shikoku Island in western Japan. The continuous monitoring has been carried out at Tokushima, Japan. From March 23, 2011 the fission product I-131 was observed. The radioisotopes Cs-134 and Cs-137 were also observed in the beginning of April. During the course of our continuous monitoring, we were also able to observe the seasonal Asian Dust (yellow dust) in the beginning of May at the same time. We found that those specific radioactivities of I-131, Cs-134 and Cs-137 fission products decrease drastically only for the period of Asian Dust, however, those levels of naturally occurring nuclides do not vary well.

1P02 Note on the dependence of radioactivity ratios of some nuclides on the sampling places

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Radioactive materials were released from nuclear reactors of Fukushima Daiichi Nuclear Power Station to the air as a result of earthquake. Main released radioactive materials were inferred high volatility or these daughter nuclides. These materials were diffused in large area of Fukushima and deposited in the soil. In this work, we examined the dependence of diffusion and precipitation by the investigated relative activity of some nuclides. We estimated the stop time of nuclear reactor by the compared iodine isotope ratios of the ambient area and these of nuclear reactor.

Soil samples of 5-cm layer from soil surface were collected by a cylindrical stainless steel apparatus (i.d.15/7inch) and measured soil samples by a high purity Ge detector coupled with a multichannel pulse height analyzer. Radio-nuclides in the air were measured by a high purity Ge detector in Tohoku University.

It was found that the activity ratios of Cs isotopes, ¹³⁴Cs, ¹³⁶Cs, and ¹³⁷Cs, are about the same in all places whereas the ratio between different elements, for example Cs and I, are deviated place to place. In Tohoku University, radio-nuclides of ¹³¹I and ¹³³I were measured and we presumed stop time of nuclear reactor by these nuclides ratio. A detailed discussion will be given in the presentation.

1P03 Environmental radioactivity monitoring at Kakuma campus of Kanazawa university after the accident of Fukushima nuclear power plant

Nakai, M., Kimura, H., Nagamura, Y., Nakanishi, T., Uesugi, M., Yokoyama, A. (Kanazawa Univ.)

The Nuclear power plant accident at Fukushima caused by the earthquake and tsunami disaster in March, 2011 led the public to fear the considerable-range contamination with radioactivity. For the reason, we started environmental monitoring on radioactivity just outside of

the RI facility in the Kakuma campus of Kanazawa Univ. A high-volume dust sampler (SHIBATA, HV-1000) was used to collect air dust onto a filter and rain water was collected in a plastic container placed on the rooftop of the facility as well. The samples were assayed by gamma-ray spectrometer with germanium detectors to obtain the concentrations of I-131, Cs-134, and Cs-137. The data were compared with those measured in the other areas in Japan to discuss how the activities diffused and migrated from the plant to many places in Japan. The peaking dates of the activities coincided with those by a trajectory calculation of air from Fukushima. The trend was confirmed for the other data in other areas.

1P04 Study on radioactive fallout from Fukushima nuclear accident by plant samples using an Imaging Plate System

Minowa, H. (The Jikei University School of Medicine)

The radioactive fallout from the Fukushima nuclear accident was investigated by the radiation images of plant samples using an Imaging Plate System. Plant samples exposed by an imaging plate BASIII 2040 (Fujifilm, Japan) in overnight to one week, and radiation images were read by Typhoon FLA7000 (GE Healthcare Japan Corp.). Identifying and quantitative analysis of radionuclides were measured by Auto Well Gamma System ARC-380CL (ALOKA Co. Ltd., Tokyo, Japan). In the cross-sectional images of the bamboo shoot, the radioactive material is shown in heterogeneous distribution, it was found that it concentrated on the tip of the edible portion, and thin skin. These radionuclides were identified as ^{137}Cs , ^{134}Cs , and ^{40}K . ^{40}K is a natural radionuclide, on the other hand ^{137}Cs and ^{134}Cs would be derived from the accident of the Fukushima Daiichi Nuclear Power Plant. A high concentration of ^{134}Cs was shown at the distance of 150mm from the base of the bamboo shoot by cross-sectional cutting into the width of about 1mm. It was estimated about 1kBq of ^{134}Cs would be included in about 400g (wet weight) of this one bamboo shoot in an edible part. Imaging data suggests that the contamination of radioactive cesium in this bamboo shoot was caused not by the extraneous attachment but by the absorption from roots. Because bamboo is gather water from extensive area, bamboo shoot concentrates the radioactive material contained in the rain even at low concentrations of radioactive materials in soil.

1P05 Behavior in water system of iodine-129 from Fukushima Daiichi nuclear disaster

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Because determination of ^{129}I by Accelerator Mass Spectrometry is a very high sensitivity, measuring the concentration of ^{129}I in tap water shows clearly the impact of Fukushima Daiichi nuclear accident. So we examined the range of effects by the accident and natural dilution of ^{129}I by the aqueous flow, by measuring the concentration of ^{129}I in tap water samples taken in Tokyo, Nagano, Aomori, and Ehime.

The concentration of ^{129}I in tap water taken in Nagano, Aomori, and Ehime were lower than 2.69×10^7 atoms/L which that of Tokyo before the accident was, while the concentration of ^{129}I in tap water taken in Tokyo in April

2011 was 1.20×10^8 atoms/L. This was considered as the impact of the accident. The concentration of ^{129}I in samples taken in Tokyo in May and in July 2011 was 6.55×10^7 atoms/L and 5.91×10^7 atoms/L, respectively. The concentration of ^{129}I has been decreased.

1P06 Measurements of the Fukushima nuclear power plants accident origin ^{137}Cs and ^{134}Cs in soil at remote place

Matsuzaki, H., Yamagata, T., Honda, M*, Tokuyama, H., Miyake, Y., Sunada, N. (Tokyo Univ., *Nihon Univ.)

Radioactive materials emitted from the Fukushima Dai-ichi nuclear power plant accident were found to be distributed to a remote places by the air stream due to the climate condition. Detailed distribution of radioactive materials even in remote places has to be measured to assess the true effect of the Fukushima Dai-ichi accident. We collected several soil samples from top 5cm layer in Kanagawa prefecture and measured ^{137}Cs and ^{134}Cs by a germanium semi-conductor detector. As an example of the results, cumulative radioactivity by ^{137}Cs and ^{134}Cs were distributed between 0.08-0.8 Bq/cm² and 0.06-0.7 Bq/cm², respectively, in Tsurumi district, Yokohama city.

1P07 Deposition flux of ^{137}Cs at Tokyo

Yamagata, T., Tsujita, K., Hirano, Y., Harada, N., Hasegawa, A., Honda, M. (Coll. Humanities and Sci., Nihon University), Inoue, K. (Grad. Sch. of Integ. Basic Sci., Nihon University), Nagai, H. (Coll. Humanities and Sci., Nihon University)

A large amount of radionuclides were discharged from Fukushima Daiichi Nuclear Power Plant by the accident caused by huge earthquake. To evaluate the amount of environmental radioactivity, the gamma ray emission nuclides such as ^{137}Cs , ^{134}Cs and ^{131}I in rainwater, tap water and aerosols were investigated from the accident at Tokyo. ^{131}I that has short half-life (8days) should be decay and extinct to a few months. The long life nuclide of ^{129}I would be used for the proxy of ^{131}I to the future. So we measured the concentration of ^{129}I in the rainwater and the tap water by Accelerator Mass Spectrometry.

The concentrations of ^{137}Cs and ^{134}Cs in the atmosphere were dramatically increased to 500 mBq m⁻³ and exponentially decreased with a environmental half-life of 9 days after the accident. The activity ratio of $^{137}\text{Cs}/^{134}\text{Cs}$ in the atmosphere was mostly constant of 1.2. The concentration of ^{129}I in the rainwater and the tap water were $(43-635) \times 10^7$ atoms kg⁻¹ and $(2.9-76) \times 10^7$ atoms kg⁻¹, respectively. The atomic ratio of $^{131}\text{I}/^{129}\text{I}$ was decreased exponentially with a half-life of 6.4 days and 7.1 days.

1P08 The secular change of radioactivity's concentration in airdust from Fukushima Daiichi Plant at Osaka, Nagoya and Ibaraki.

Cho, S.¹, Nakamura, K.¹, Shiohara, N.¹, Yashuda, Y.¹, Shinohara, A.¹, Takahashi, N.¹, Saito, T.², Oda, H.³, Ikeda, A.⁴, Kita, K.⁵, Kasahara, R.⁵ (¹School of Sci., Osaka Univ., ²Dept. Administration of Safty & Hygiene, Osaka, Univ., ³Center for Chronological Res., Nagoya Univ., ⁴Technical Center of Nagoya Univ., ⁵College of Sci., Ibaraki Univ.)

The polluted air dust by radioactive waste from Fukushima No. 1 nuclear power plant have been measured from March 2011 onward at Osaka (Toyonaka), Aichi

(Nagoya) and Ibaraki (Mito and Hitachi). Radioactivity of ^{131}I , ^{134}Cs , ^{136}Cs and ^{137}Cs were measured by HPGe γ -ray spectrometer for the 365, 605, 819 and 662 keV γ -ray, respectively. Iodine-131 had been first detected from March 18 in Nagoya, and ^{134}Cs and ^{137}Cs began to be observed after 10 days. Concentrations of ^{131}I , ^{134}Cs and ^{137}Cs in the air were in the range of 0 to 14, 0 to 18 and 0 to 20 mBq/m³, respectively, with two peaks in April 6 and 17. These peaks may be suggested by the existence of high concentration of polluted air mass which is rotated around the earth.

1P09 Activity measurement of airborne dust in Osaka Sennan area

Oki, Y., Osada, N., Yamasaki, K., Shibata, S. (Kyoto Univ.)

Radioactive materials released from the accident site of Fukushima Daiichi Nuclear Power Plant spread far and wide as airborne dust. In this work, the airborne dust was collected in Sennan area in Osaka, and its radioactivity was measured. The dust was sampled everyday at Kyoto University Research Reactor Institute in Kumatori-cho, Osaka on a glass fiber air sampling filter using a high volume air sampler. The activity was measured with Ge detector systems. The principal radionuclides found on the filter were ^{131}I , ^{134}Cs and ^{137}Cs . Their activity concentrations in air were estimated and their fluctuations were discussed.

1P10 Environmental radioactivity measurements at Mikamine, Sendai

Kikunaga, H., Ohtsuki, T., Ishikawa, T., Kashiwagi, S., Shibasaki, Y., Suzuki, K., Sugawara, Y., Takahashi, K., Takahashi, S., Nagasawa, I., Nanbu, K., Hiraga, M., Fujimura, H., Matsuda, K., Muto, T., Miyahara, F., Miyabe, M., Yamazaki, H. (Research Center Elec. Photon Sci. Tohoku Univ.)

The Fukushima 1 Nuclear Power Plant suffered major damage from the 2011 off the Pacific coast of Tohoku Earthquake and subsequent tsunami on March 11, 2011 and released various radionuclides. Monitoring of environmental radioactivity should provide important information on the behavior of the radionuclides. In this presentation, the monitoring results on the concentration of radioactivity in the air and soil and the radiation dose rate are shown. In addition, the Electron Linear Accelerator at Research Center for Electron Photon Science, Tohoku University also suffered damage from the earthquake. The state and the reconstruction plan of the accelerator will be presented.

1P11 Pollution by radioactive iodine and radioactive cesium in fruit fields

Ohtsuki, T., Kikunaga, K. (Tohoku Univ.), Izumi, Y. (Nikkannken Co.), Kirishima, A., Satou, N. (Tohoku Univ.), Muramatsu, Y., Ohno, T. (Gakusyuin Univ.), Sato, M., Abe, M., Abe, K. (Research Institute of Fruit)

A lot of radioactive materials were spread out by the accident of the Fukushima Daiichi nuclear power station caused by the tsunami accompanying earthquake in the area of Tohoku district-Pacific Ocean, in 2011/3/11. In early stage, short half-life nuclides, such as ^{132}Te , ^{131}I , ^{136}Cs , as well as ^{134}C , ^{137}Cs were observed in fields. Now ^{134}C and ^{137}Cs which are comparatively long half-life became

dominant observing nuclides. The nuclear power plant accident brought serious damages in many directions, especially, the influence of the field pollution by these nuclides is great in the agriculture, and the quick actions and the measures are needed. The influence of ^{134}C and ^{137}Cs to the fruit tree were investigated in cooperation with Fukushima agricultural center, Institute of Fruit Tree Science. We report several facts of the transfer and translocation mechanism of ^{134}C and ^{137}Cs in fruit trees.

1P12 Concentration and size-distribution of airborne radionuclides at Nagano City after the accident at the Fukushima I nuclear power plant

Muramatsu, H., Kawasumi, K., Kondo, T. (Fac. of Edu., Shinshu Univ.)

Concentrations of radionuclides released from the Fukushima I nuclear power plant have been measured for surface air, rain and soil at Nagano City (36°39'N, 138°12'E). By using γ -ray spectrometry of collected samples we have detected and determined fission products of ^{131}I , ^{132}Te , ^{134}Cs and ^{137}Cs . Concentrations of relatively short-lived nuclides, ^{131}I and ^{132}Te , in surface air rapidly decreased with increasing elapsed time from the accident mainly due to radioactive decay and dilution.

Concentrations of long-lived nuclides, ^{134}Cs and ^{137}Cs , in wet precipitation (rain and snow) gradually decreased due to diffusion and dilution of airborne species in air, and due to so-called *washout* every rainfall.

The particle size distribution of radioactive aerosols has been observed using the Andersen-type classifier combined with the high volume air sampler. Aerosol particles were classified and collected on four glass fiber filters, and on a back-up filter for aerosols smaller than 1.1 μm in diameter. About 80% of collected activities was found in the back-up filter, which means that the majority of radioactivity would attached to aerosols with the diameters less than 1.1 μm . Using three data corresponding to particle-size ranges of 3.3-7.0, 2.0-3.3 and 1.1-2.0 μm , Activity Median Aerodynamic Diameter (AMAD) was estimated under the assumption that the particle size distribution of radioactive aerosols in surface air is in a lognormal and unimodal distribution. Estimated AMADs of ^{131}I -, ^{134}Cs - and ^{137}Cs -aerosols were ranging from 0.56 to 0.60 μm , which were greater than that of ^7Be -aerosols, about 0.2 μm .

1A01 Surface-ionization of lanthanides for the measurement of the first ionization potential of Lr

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The first ionization potential (IP) of the last actinide, lawrencium (Lr), is predicted to be lower than those of other actinides. To determine the IP of Lr based on the surface ionization comparison technique, we are developing a surface-ionization-type ion-source coupled to a gas-jet transport system at the Isotope Separator On-Line at the JAEA tandem accelerator facility. In the present study, we measured ionization efficiencies of lanthanides as a function of ion-source temperature to examine the applicability of our apparatus to the IP measurement. It was found that the obtained ionization efficiency of each lanthanide element is affected by an ionization potential as well as a boiling point.

1A02 Determination of redox potential of mendelevium (Md)

Toyoshima, A.¹, Li, Z. J.¹, Asai, M.¹, Sato, N.¹, Sato, T. K.¹, Ooe, K.¹, Kikuchi, T.¹, Kaneya, Y.¹, Kitatsuji, Y.¹, Tsukada, K.¹, Nagame, Y.¹, Schädel, M.^{1,2}, Kasamatsu, Y.³, Kogama, Y.³, Shinohara, A.³, Haba, H.⁴, Even, J.⁵ (¹JAEA, ²GSI, ³Osaka Univ. ⁴RIKEN, ⁵Universität Mainz)

Redox potential of mendelevium (Md) was determined with flow electrolytic chromatography. Mendelevium-255 with a half-life of 27 min was produced in the ²⁴⁸Cm(¹¹B, 4n) reaction at the JAEA tandem accelerator. Reaction products transported by a KCl/He gas-jet method were collected on a chemistry apparatus. After removing KCl with HDEHP column chromatography, elution behavior of Md in 0.10 M HCl was investigated with a flow electrolytic column apparatus at the applied potentials between -0.3 and -0.6 V vs. a Ag/AgCl reference electrode. At -0.3 V, elution behavior of Md was the same as that of ²⁵⁰Bk³⁺, showing that the stable Md³⁺ was not reduced to the divalent state. At -0.6 V, elution of Md was quite similar to that of Sr²⁺, demonstrating that Md³⁺ was successfully reduced to Md²⁺. From the behavior of Md against the applied potentials, we evaluated its redox potential to be -0.40±0.03 V.

1A03 Development of a liquid scintillation detection system for aqueous chemistry of seaborgium

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The purpose of our study is to develop an on-line liquid scintillation detection system coupled to an ion-exchange or a liquid-liquid extraction apparatus for aqueous chemistry experiments of element 106, seaborgium. In this work, we developed a liquid scintillation detector with α/β discrimination for on-line measurements. Energy resolution and detection efficiency in α -particle detection was measured using the nuclide ²²⁶Ra and ²⁴¹Am, respectively. Events derived from spontaneous fission were observed using the nuclide ²⁵²Cf. A procedure of the on-line measurement is as follows: the emulsifier scintillator was introduced into the cell, and then aqueous solution sample was injected with mixing by using an electromagnetic stirrer. Measurement was started after homogeneously mixing of the solution in the cell. After the measurement, solution in the cell was drained. This procedure was automated and repeated. This detection system was applied to on-line measurement of ²¹³Fr ($T_{1/2} = 34.6$ s) which was produced by ²⁰⁹Bi(¹⁶O,4n)²²¹Pa reaction using the AVF cyclotron at the Research Center for Nuclear Physics.

1A04 High-resolution alpha fine-structure spectroscopy for studying nuclear structure of heavy nuclei

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We have developed high-resolution alpha fine-structure spectroscopy for studying nuclear structure of

short-lived heavy actinide nuclei. It has been demonstrated that spin-parities and single-particle configurations of the ground state of a parent nucleus as well as excited states in a daughter nucleus can be determined only through the measured alpha singles spectrum. To achieve a good alpha energy resolution of ~11 keV, conditions to make a thin alpha source have been optimized. Using this technique, we have identified proton single-particle configurations of Lr-255g, 255m, 257, and 259 for the first time.

1B01 A Relationship between Water Purifying Effect and Structure of Iron Containing Soda-lime Silicate Glass

Iwanuma, Jun; Kubuki, Shiro; Akiyama, Kazuhiko (Tokyo Metropolitan Univ.), Nishida, Tetsuaki (Kinki Univ.)

Chemical Oxygen Demand (COD) achievement ratio, which is an index of the water contamination of closed water systems like lake or pond, is lower than that of open water systems like river and ocean, because contaminants tend to accumulate at the bottom. Current water purifying methods have problems with cross-contamination by chemicals *etc.* In this study, a relationship between water purifying effect and the local structure of iron containing soda-lime silicate glass (xNCFS glass), prepared by conventional melt-quench method at 1400 °C for an hour, was investigated by means of COD manganese method (K0102 17), Powder X-ray diffraction, and ⁵⁷Fe-Mössbauer spectroscopy. Two paramagnetic doublets due to Fe²⁺ and Fe³⁺ were observed in Mössbauer spectra of 10 and 30NCFS glass, and only a paramagnetic doublet due to Fe³⁺ was confirmed in 50NCFS glass. In the case 50NCFS glass, the COD value was significantly decreased from 276 to 98.7 mg/L after 3days. On the other hand, in the case 10 and 30NCFS glass, the COD value was significantly decreased from 236 to 64.5 and from 234 to 67.5 mg/L after 7days, respectively. It is concluded that 50NCFS glass decreases COD most effectively.

1B02 Electrical Conductivity and Local Structure of Chemically-modified Barium Iron Vanadate Glass

Masuda, H., Kubuki, S., Akiyama, K. (Tokyo metropolitan Univ.), Nishida, T. (kinki Univ.)

A relationship between local structure and electric conductivity (σ) of manganese substituted *NTA* glassTM with a composition of 20BaO · (70-x)V₂O₅ · 10Fe₂O₃ · xMnO₂ (xMn*NTA*) was investigated by means of ⁵⁷Fe-Mössbauer spectroscopy, and dc four-probe method.

Mössbauer spectrum of 10Mn*NTA* glass consists of a paramagnetic doublet with the isomer shift (δ) and quadrupole splitting value (Δ) of 0.41 mm s⁻¹ and 0.76 mm s⁻¹ indicating that Fe atom forms distorted Fe^{III}O₄ tetrahedra. A drastic decrease in Δ value from 0.76 to 0.49 mm s⁻¹ was observed after the isothermal annealing. A marked increase in σ was observed from (4.7±3.8)×10⁻⁷ to (1.4±0.3)×10⁻² S cm⁻¹ when 10Mn*NTA* was isothermally annealed at 500 °C for 1000 min.

An increase in σ accompanying decrease in Δ value was also confirmed after the annealing of phosphorous substituted *NTA* glassTM with a composition of 20BaO · (70-x)V₂O₅ · 10Fe₂O₃ · xP₂O₅. These results indicate that the increase in σ value is closely related to decrease in Δ value of vanadate glass.

1B03 Mössbauer spectroscopic study of incorporations of organic molecules into intermolecular voids in the mixed-valence trinuclear iron(III, III, II) pentafluorobenzoate complex

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The mixed-valence trinuclear iron (III, III, II) carboxylate complexes have received intensive attention from many chemists in the field of Mössbauer spectroscopy, in which the valence states of the 3 iron ions are trapped or detrapped (averaged) depending on various conditions. It was proved in our previous Mössbauer studies using pentafluorobenzoate as carboxylate that the valences of three iron ions are trapped at room temperature for $\text{Fe}^{\text{III}}_2\text{Fe}^{\text{II}}\text{O}(\text{C}_6\text{F}_5\text{COO})_6(\text{C}_5\text{H}_5\text{N})_3$ (abbr. *F5*) while they are detrapped at room temperature for $\text{Fe}^{\text{III}}_2\text{Fe}^{\text{II}}\text{O}(\text{C}_6\text{F}_5\text{COO})_6(\text{C}_5\text{H}_5\text{N})_3\cdot\text{CH}_2\text{Cl}_2$ (abbr. *F5.CH₂Cl₂*), CH_2Cl_2 being here a solvate molecule in crystalline solid. Our present study showed that Mössbauer spectra at room temperature of *F5* exposed to vapor of organic compounds, such as CH_3CN , CH_2Cl_2 , and C_6H_6 , were transformed from an iron-valence trapped type to a detrapped type. To the contrary, such a transformation was not observed for the case of exposing to $\text{C}_6\text{H}_5\text{CH}(\text{CH}_3)_2$ with a relatively large molecular size. This finding allows us to presume tentatively that some gaseous organic molecules (*M*) should incorporate into the intermolecular void with a nano-meter size of *F5* crystals, subsequently forming the solvate crystal *F5.M*.

1B04 Syntheses and Mössbauer spectroscopic study of mixed-valence hetero-trimetallic pentafluorobenzoate complexes containing iron ions

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In our previous study, ⁵⁷Fe-Mössbauer spectra were measured for mixed-valence trinuclear iron (III, III, II) pentafluorobenzoate complexes, $\text{Fe}^{\text{III}}_2\text{Fe}^{\text{II}}\text{O}(\text{C}_6\text{F}_5\text{COO})_6(\text{C}_5\text{H}_5\text{N})_3\cdot\text{CH}_2\text{Cl}_2$, where CH_2Cl_2 is a solvate molecule in crystalline solid. It was shown that all iron ions should be in a fully detrapped valence state of +2.7, i.e., an average value of 2Fe^{3+} and Fe^{2+} , to which one doublet was assigned at 300 K while the trapped valence states of Fe^{3+} and Fe^{2+} were separately observed at 5 K, and that the transition temperature from the trapped state to the detrapped one was estimated to range from 70 to 100 K. In the present work, one of Fe^{3+} ions of the above complex being substituted with Cr^{3+} , $\text{Cr}^{\text{III}}\text{Fe}^{\text{III}}\text{Fe}^{\text{II}}\text{O}(\text{C}_6\text{F}_5\text{COO})_6(\text{C}_5\text{H}_5\text{N})_3\cdot\text{CH}_2\text{Cl}_2$, was synthesized as a novel compound. The ⁵⁷Fe-Mössbauer spectra were measured at temperatures from 78 K to 300 K, revealing that Fe^{3+} and Fe^{2+} were separately observed in the trapped valence state at 78K while there was found only one component at 300 K, ascribable to $\text{Fe}^{2.5+}$ in the detrapped valence state. It is the first observation that valence detrapping between Fe^{3+} and Fe^{2+} ions should occur in a mixed-valence hetero-trimetallic carboxylate complex.

1B05 Studies on hot atom chemical behavior of energetic ions in solids(XXVI) ~ Dependence of impurity

concentration on hydrogen isotope retention in carbon and oxygen containing boron films

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Boronization has been considered to be applied as one of the most effective techniques for first wall conditioning in D-T fusion reactors. Boron is easily bound to impurities such as carbon and oxygen, and the boron film contaminated by them impurities will be formed. It is considered that tritium will be trapped not only by boron but also the impurities in the boron film. In the present study, the boron films contaminated by both of carbon and oxygen were prepared and the impurity effects of carbon and oxygen on deuterium retention behavior were studied.

The boron films were prepared by the Plasma-assisted Chemical Vapor Deposition method and then the samples were removed residual hydrogen by heating up to 1173 K. Thereafter, deuterium ion (D_2^+) was implanted into the sample. Before and after the D_2^+ implantation, the XPS measurements were carried out. The TDS measurements were performed from room temperature up to 1173 K.

Three D_2 desorption stages were appeared around 550 K, 750K and 900 K in the D_2 TDS spectra, attributing to the desorption processes of deuterium trapped as B-D-B bond, B-D bond and B-O-D and/or B-C-D bonds, respectively. The amount of C bound to B as C-B bond was increased as the carbon concentration increased and that of O as free oxygen was also increased with increasing the oxygen concentration in boron films, although that as C-B bond for the 20% carbon-contained boron films was little increased. These results indicate that hydrogen isotopes are trapped as B-C-D bond in lower carbon concentration when oxygen co-exists with carbon in boron films.

1B06 Studies on hot atom chemical behavior of energetic ions in solids(XXVII)-Elucidation of deuterium retention behaviors in only D_2^+ irradiated tungsten

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Tungsten is considered to be used as plasma facing materials in D-T fusion reactors. Irradiation defects are formed in tungsten when high energy particles escape from plasma. Especially, elucidation of retention behavior of hydrogen isotopes including tritium is necessary with respect to tritium inventory. In this study, D_2^+ irradiation was performed as a function of D^+ fluence in the range of $(0.03\text{-}1.8) \times 10^{22} \text{D}^+ \text{m}^{-2}$. The deuterium retention behavior in tungsten was investigated by means of Thermal Desorption Spectroscopy (TDS) and Glow Discharge – Optical Emission Spectroscopy (GD-OES), and irradiation defects were observed by Transmission Electron Microscope (TEM).

The D_2 TDS spectrum was consisted of 3 deuterium desorption stages around 400 K, 550 K and 650 K. These peaks were attributed to the desorption of deuterium adsorbed on the surface and/or trapped by dislocation loop (Peak 1), that retained in vacancy (Peak 2) and that migrated toward the bulk (Peak 3), respectively. Above $1.0 \times 10^{22} \text{D}^+$

m², the deuterium retention as Peak 1 was saturated and that as Peak 3 was increased, indicating that deuterium would diffuse toward the bulk after almost all the surface trapping sites were saturated by deuterium. The TEM observations showed that the density of dislocation loops was increased as the D⁺ fluence increased. Increasing the amount of dislocation loops indicated that much more vacancies were introduced in tungsten. The GD-OES measurements showed that the D depth profile was shifted toward the bulk as the D₂⁺ fluence increased. These results indicated that deuterium was diffused toward the surface by trapping and detrapping by damages, which induce the higher desorption temperature of Peak 3. It was revealed that the peak shift and the amount of D retention have a correlation to the density of defects.

1B07 Studies on hot atom chemical behavior of energetic ions in solids (XXVIII) - Elucidation of chemical release process for tritium produced in thermal neutron-irradiated Lithium titanate –

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In blanket systems for D-T fusion reactors, it will be considered that lithium ceramics are introduced to breed tritium by the reactions of ⁶Li(n,α)T and ⁷Li(n,n'α)T. Li₂TiO₃ is considered to be used as a solid tritium breeding material due to its low reactivity and high tritium recovery performance. In order to realize an effective tritium fuel cycle for fusion reactors, the elucidation of tritium release behavior is required. Therefore, tritium release processes in neutron-irradiated Li₂TiO₃ were kinetically elucidated by means of Thermal Desorption Spectroscopy (TDS). Li₂TiO₃ powder purchased from KAKEN Co., Ltd was irradiated by thermal neutrons with the fluence of 3.3 x 10¹⁵ n cm⁻² at the Kyoto University Research Reactor Institute (KURRI). The TDS measurements were performed with the heating rates of 5-30 K min⁻¹ in the temperature range of 300-1373 K, and isothermal heating was also applied in the temperature range of 500-600 K to clarify tritium desorption dynamics. It was found that 98 % of tritium was desorbed as HTO and the rest was desorbed as HT. The desorption stage around 800 K of the HTO-TDS spectrum was attributed to the desorption of tritium retained in the irradiation defects. The heating spectra were well fitted by a diffusion equation assumed sphere indicating that the rate-determining step of tritium release is the diffusion of tritium in Li₂TiO₃. The tritium release processes are discussed in detail by simulating the isothermal heating spectrum and calculation of activation energy from heating rate dependence measurements.

1B08 Studies on hot atom chemical behavior of energetic ions in solids (XXIX) –Clarification for influences of chemical states of stainless steel oxide layer on retention behaviors of hydrogen isotopes-

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In fusion devices, it is considered that stainless steel (SS316) will be widely used as component materials such as a vacuum vessel, cooling pipes and so on. An oxide layer is formed on the surface of stainless steel and hydrogen isotopes including tritium is trapped and retained in the oxide layer. It is seen that the chemical states of the oxide layer is controlled by the oxidation temperatures. Clarification of correlation between chemical states of the oxide layer and tritium retention behavior is important from the viewpoint of tritium retention control. In this study, influences of chemical states of the oxide layer on retention behavior of hydrogen isotopes were studied.

Following to the pretreatment, the stainless steel samples were exposed to Ar/O₂ gases in order to form the oxide layers at R.T., 473 K or 673 K. Thereafter, these samples were exposed to D₂ gas at 573 K. The deuterium desorption behavior was observed by Thermal Desorption Spectroscopy (TDS) in the temperature range of R.T.-1173 K. The chemical states of the surface oxide layer were investigated by X-ray photoelectron Spectroscopy (XPS)

The TDS results showed that the amounts of D₂ desorption increased with increasing the oxidation temperature, although D₂O desorption was independent of the oxidation temperature. The D₂ desorption stages were found around 600, 700 and 800 K, attributing to the releases of deuterium adsorbed on the surface, trapped by the oxide layer and retained in the bulk, respectively. According to the XPS measurements, the major chemical state of the surface oxide layer was iron oxides and the deuterium desorption temperature was consistent with the decomposition temperature of iron oxides, suggesting that the chemical states of iron oxide would affect the deuterium retention in the oxide layer.

1B09 In-beam Mössbauer spectroscopy of ⁵⁷Mn implanted into lithium hydride

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A study of ⁵⁷Fe in-beam Mössbauer spectroscopy using ⁵⁷Mn ion beams provides direct information on exotic state of unstable nuclear probes and on electronic properties of the surrounding materials. Recently, our detection system was successfully improved to obtain in-beam Mössbauer spectra with very large S/N ratio using Heavy Ion Medical Accelerator in Chiba (HIMAC) of NIRS. Binary compounds of Fe or Mn atom and H atom have not been synthesized, and a stable ternary compound Mg₂FeH₆ has been known. In the present work, we investigated the chemical and physical behaviors of ⁵⁷Fe arising from ⁵⁷Mn in a lithium hydride, which is an ionic solid having a very simple rock salt type crystal structure. The ⁵⁷Mn secondary beam was produced through a nuclear projectile fragmentation process of the ⁵⁸Fe beam with a 27mm-thick Be plate as a production target. The LiH polycrystalline sample, which was purchased from Wako Pure Chemical Industries, was mounted on a BN heater. The sample thickness was around 3 mm. The temperature dependence of Mössbauer spectra of ⁵⁷Mn/⁵⁷Fe in LiH was obtained in the range from room temperature to 823 K. The obtained spectra were fitted with the combination of a singlet and

two doublets. The singlet peak of which the isomer shift is $\delta = 0.29(3)$ mm/s is assigned to an Fe atom surrounded by six H atoms, which substitutes Li atom in LiH crystal. Because the difference between the electronegativities of Fe and H is comparatively small, the covalent bond is considered to be formed. Density functional calculation with cluster models is in progress to make assignments for the other components.

1B10 Dilute magnetism and Mössbauer spectra in Fe and Co co-doped ZrO₂

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We investigate the effects of Fe and Co co-doping on the magnetic and electronic properties of ZrO₂ ceramics prepared by a sol-gel method, and study their dependence on the annealing temperature. Dilute Fe and Co co-doping into ZrO₂ exhibits ferromagnetic behavior at room temperature for annealing temperatures above 900 °C, accompanying the phase transition from tetragonal to monoclinic structure in ZrO₂. The electronic structures are studied by x-ray absorption spectroscopy and Mössbauer spectroscopy, which suggest that the Fe³⁺ and Co²⁺/Co³⁺ mixing states are dominant in Fe and Co co-doped ZrO₂. In Mössbauer spectra, as the annealing temperature increases to 900 °C, sextet peaks appear in the co-doped case, and these are enhanced at 1000 °C. The appearance of sextet peaks reveals the magnetic ordering in Fe ions and an internal magnetic field of 40 T. Broad sextet line shapes and hyperfine fields originate from the formation of nanoparticles and super-paramagnetic contributions from CoFe₂O₄ (Co-ferrite) like compounds. As the internal magnetic field in bulk CoFe₂O₄ is about 50 T, the line shapes can be explained by the size effect in the segregation of the Co-ferrite-based nanoparticles.

1B11 Syntheses of Mixed Crystals of Assembled Iron Complexes Bridged by 1,3-Bis(4-pyridyl)propane and Their Spin States (III)

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Self-assembled coordination polymers containing transition metal ions and organic bridging ligands have attracted intensive interests because of their potential abilities for selective inclusion and transformation of ions and molecules. In the present study, we discussed the structure of the mixed crystals of zinc and cobalt ion for the assembled complexes bridged by bpp, and then we discussed the spin state of the mixed crystals of iron ion with zinc and cobalt ions for the assembled complexes bridged by bpp.

Mixed crystals were obtained by diffusion method. Zinc and cobalt ions were mixed in the synthetic process. From the solution, red and blue crystals were obtained. Powder X-ray diffraction pattern showed that the red crystals are in the 2D interpenetrated structure, and the structure of blue crystals is similar to that of Zn(NCS)₂(bpp). ICP-OES revealed that red crystals have relatively larger cobalt content, while blue crystals have larger zinc content.

Iron, zinc, and cobalt ions were mixed in the synthetic process. From the solution, red and blue crystals were obtained. Powder X-ray diffraction pattern showed that the red crystals are in the 2D interpenetrated structure, and the

structure of blue crystals is similar to that of Zn(NCS)₂(bpp). ICP-OES revealed that red crystals have relatively larger cobalt content, while blue crystals have larger zinc content. And ICP-OES revealed that the content of iron is relatively low both in red and blue crystals.

⁵⁷Fe Mössbauer spectroscopy revealed that the blue mixed crystals have a trend to show Fe(II) low-spin state, while red mixed crystals show Fe(II) high-spin state.

1B12 Strong affinity between dilute Al and In impurities doped in ZnO

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The time-differential perturbed angular correlation method was applied to a study of local fields at the ¹¹¹Cd(←¹¹¹In) probe nucleus in Al-doped ZnO. In our previous TDPAC studies, we found that local fields at the probe nucleus in In-doped and Al-doped ZnO are distinctly different from that for undoped ZnO. Especially for the Al-doped ZnO, in addition, the doping effect on the ¹¹¹Cd(←¹¹¹In) probe is prominent even at as low Al concentration as 500 ppm. This experimental result implies that there is a strong attractive force between Al ion(s) and the ¹¹¹Cd(←¹¹¹In) probe. By means of the TDPAC method, in this work, we investigated the dilute limit of Al concentration at which their strong affinity between Al and the ¹¹¹Cd(←¹¹¹In) probe presumed above can be observed. We found that in Al-doped ZnO the ¹¹¹Cd(←¹¹¹In) probe is perturbed by the local field produced by the doped Al at the Al concentration as dilute as 0.1 ppm.

1B13 Anomalous electric conductivity observed for In-doped ZnO

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Zinc oxide (ZnO) is an intrinsic *n*-type semiconductor, and its conductivity varies with the presence of dilute impurities. It is hence important to investigate how impurity ions function in ZnO matrix. In our previous work, we applied the time-differential perturbed angular correlation (TDPAC) method with the ¹¹¹Cd(←¹¹¹In) probe to a study of local fields in 0.5 at.% In-doped ZnO, and found that the density of conduction electrons at the probe site is lower than that for undoped ZnO. In the present study, therefore, we have measured electric resistivity of the In-doped and undoped ZnO samples in order to compare the density of conduction electrons in a macroscopic scale. The resistivity for the 0.5 at.% In-doped ZnO was three-to-four orders of magnitude greater than that for the undoped one, which is inconsistent with general understanding that donor impurities provide conduction electrons. The local structure in the vicinity of the probe in ZnO matrix is discussed based on the resistivity as well as the electric field gradients at the probe nucleus.

1B14 Electronic Structures and Photophysical Properties of Nitridotechnetium(V) and Rhenium(V) Complexes Showing Reversible Interconversions

between Five- and Six-coordinate Structures

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A single chemical imaging agent applicable to both radio- and optical methods has attracted much attention. In this circumstance, the fundamental photoluminescent properties of technetium and rhenium complexes are worth studying in detail for development of a radio- and photo-active agent based on a Group 7 element. In this study, novel five- and six-coordinate technetium(V) and rhenium(V) complexes were synthesized and their photoluminescence properties and electronic structures were investigated. Furthermore, we report the reversible reaction between five- and six-coordinated complexes.

It is found that the emission peak of six-coordinate rhenium complex shifted to the shorter wavelength than that of six-coordinate technetium one. In the case of five-coordinated structure, the technetium complex showed emission peak at shorter wavelength than rhenium one.

In both of technetium and rhenium complexes, the interconversion between six-coordinate complex with methanol and five-coordinate complex was occurred. Furthermore, in the case of rhenium complex, the substitution reactions between the six-coordinate complexes with methanol and acetone were observed in the solid state at room temperature. On the other hand, in the case of technetium, the five-coordinate complex was obtained from the reaction of the methanol coordinate complex in the acetone vapor in the solid state.

1B15 Formation of colloidal ⁷Be species in the cooling water for magnetic horns at the J-PARC Neutrino Facility.

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At the J-PARC Neutrino Facility, ⁷Be is induced in the cooling water for magnetic horns by spallation of oxygen in water molecules. Colloid formation behavior of ⁷Be in the cooling-water was investigated in relation to the adsorptivity of ⁷Be on ion-exchange resins. After circulating the cooling water through the ion-exchange resin units for several-ten hours, a part of ⁷Be remained in the circulating water and could not be removed quickly by ion-exchange resins. Colloidal fraction of ⁷Be in the water after circulation through the ion-exchange resin units was found to be 15 to 40 % depending on the beam operations. The colloidal ⁷Be species were dissociated into soluble species by addition of dilute sulfuric acid in water. Adsorptivity of ⁷Be on cation-exchange resins also increased in acidic conditions. These results imply that adsorptivity of colloidal ⁷Be species on ion-exchange resins is lower than that of soluble ⁷Be species and it can be easily increased by adding dilute sulfuric acid in circulating water.

1B16 Mineral analysis of some commercially available Japanese tea leaves and tea infusions by

neutron-induced prompt and delayed gamma-ray analysis

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Tea is one of the most consumed beverages in the world. In Japan, the consumption of green tea is very high as compared with oolong and red teas. Determination of mineral contents of tea, especially in tea infusions is very important to elucidate the amount of the elements intake during tea consumption. In this study, we aimed to determine mineral elemental contents before and after infusion of some commercially available green teas and to determine percentage released of the elements during infusion with boiling water. Six brands of commercially available tea leaves, representing different regions of Japan, were purchased from local markets for analysis. Tea leaves before and after infusions were analyzed by PGA and INAA at JRR-3 of JAEA. Elemental contents of K, Ca, Mg, Al, S, Na, Mn, Fe, Zn, Cl, Br, B, La, Sm, Sc and Co were determined in tea leaves. Boron and S were determined by PGA. Among the determined elements the major elements are K, Ca, Mg, Al, S and Mn (concentrations ranged from 2.50 % to 500 µg/g). The elemental contents vary in tea leaves from region to region where tea is cultivated. The average extraction rates into infusion of some highly soluble elements like Cl, Br, K, Co, Na, S, Zn, B, Mg and Mn are 93, 80, 71, 60, 49, 39, 36, 16, 14 and 5% in 6 minutes extraction, respectively. Among these elements Cl, Br and K are the most soluble elements in the infusion. The high solubility of Cl, Br and K in tea infusion can be explained by the fact that these elements are present in plants in soluble forms. The present study suggests that tea infusion contains a high amount of micronutrients like K, Ca, Na, Mg and Mn which are essential minerals to human health.

1P13 Development of Si-CdTe detector array for Nuclear spectroscopy of superheavy nuclides ②

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Nuclear spectroscopy for understanding of nuclear shell-structure is one of the hot topics in nuclear physics on the superheavy nuclei. Near future, the nuclear spectroscopic study at the focal plane of GARIS or GARIS-II in RIKEN will be started. We pay attention to the CdTe detector as $\gamma(X)$ -ray detector instead of the Ge detector, because the radiation background due to neutron and $\gamma(X)$ -ray is high at RILAC in RIKEN. Thus, we developed a new Si-CdTe detector array with large active-area. Additionally, optimum condition for operating each detector was studied. In this symposium, we will report about a possibility of the Si-CdTe detector array for $\alpha(e)-\gamma$ spectroscopy of superheavy nuclei.

1P14 Preparation of ⁹⁹Mo and ¹⁸¹W radiotracers and their application for anion-exchange studies

Wada, A., Oura, Y., Haba, H., Kanaya, J. (Tokyo Metropolitan Univ., RIKEN)

In chemical studies of element 106, Sg, it is important to compare chemical properties of Sg with those of its lighter homologues, Mo and W. The chemical procedures to prepare no-carrier-added radiotracers, of ⁹⁹Mo and ¹⁸¹W,

were investigated in this work. ^{99}Mo was obtained as a fission product of a ^{252}Cf source and ^{181}W was produced by the $^{181}\text{Ta}(p, n)^{181}\text{W}$ reaction at the RIKEN AVF cyclotron. They were purified by anion-exchange method. In addition, an excitation function for the $^{181}\text{Ta}(p, n)^{181}\text{W}$ reaction was determined and compared with those in the literature and the theoretical calculations by TALYS code.

1P15 Study on cation-exchange kinetics of Zr and Hf as homologues of $_{104}\text{Rf}$ with H_2SO_4 solutions

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It was reported that the cation-exchange chromatographic behavior of Zr and Hf as homologues of elements 104, Rf, in H_2SO_4 solutions was not consistent with the distribution coefficients obtained by the batch experiment. This study was investigated on the ion-exchange kinetics which were obtained from diffusion processes of Zr and Hf on the cation-exchange resin in H_2SO_4 solutions, and was discussed about the quantitativity of chromatography. We obtained the fractional attainment of equilibrium, $U(t)$, of the carrier-free radiotracers ^{88}Zr and ^{175}Hf adsorbed on the cation-exchange resin(MCI GEL CK series, H^+ form) after a contact time, t , and then variations of $U(t)$ values in the H_2SO_4 solutions as a function of contact time within 300 seconds by the batch experiment. We discussed the dependence of functions of $U(t)$ on temperatures, concentrations of HSO_4^- and radii of resins. It is result that the good agreement between experimental data and theoretical curves which the film diffusion control is the rate-determining step in the present ion-exchange process.

1P16 Production of radioisotopes for commercial distribution

KANAYA, J., KAMBARA, T., HABA, H., TAKAHASHI, K., (RIKEN Nishina Center)
YAMADA, T., WAKITANI, Y., YAMAMOTO, S. (Japan Radioisotope Association)

In October 2007, we started a charged distribution of the purified radioisotopes (RIs) of ^{65}Zn and ^{109}Cd to the general public with the collaboration of Japan Radioisotope Association. In December 2009, we added ^{88}Y into a list of RIs available for the charged distribution. ^{65}Zn , ^{109}Cd , and ^{88}Y were produced in the $^{65}\text{Cu}(p, n)^{65}\text{Zn}$, $^{109}\text{Ag}(p, n)^{109}\text{Cd}$, and $^{88}\text{Sr}(p, n)^{88}\text{Y}$ reactions, respectively, with a 14-MeV proton beam ($\sim 20 \mu\text{A}$) from the RIKEN AVF cyclotron. After the irradiation, ^{65}Zn was chemically separated from the target material by an anion-exchange method in a hydrochloric acid system. ^{109}Cd was separated in the same method as ^{65}Zn after a AgCl precipitation. ^{88}Y was separated from the target material by a reversed-phase chromatography with the Eichrom Ln resin in a nitric acid system. The production yields, chemical impurities and specific activities of those purified RIs were evaluated using a Ge detector and an ICP-MS. The production yields of ^{65}Zn and ^{109}Cd were 0.24 and 0.080 $\text{MBq } \mu\text{A}^{-1} \text{h}^{-1}$, respectively, and the specific activities were ≥ 242.8 and $\geq 94.3 \text{ MBq } \mu\text{g}^{-1}$, respectively.

1P17 Extraction of Mo and W in oxidation states +5 and +6 with Aliquat 336

Yokokita, T.¹, Ooe, K.², Komori, Y.¹, Kikutani, Y.¹, Kino, A.¹,

Kasamatsu, Y.¹, Yoshimura, T.¹, Takahashi, N.¹, Shinohara, A.¹ (¹Grad. School of Sci., Osaka Univ., ²Advanced Science Research Center, JAEA)

We are planning to study a redox behavior of element 106, seaborgium (Sg) based on the comparison of the behavior with those of molybdenum (Mo) and tungsten (W). In this work, we studied solvent extraction behaviors of Mo(V), Mo(VI), and W(VI) with Aliquat 336 from hydrochloric acid (HCl). Mo(VI) and Mo(V) solutions were prepared by dissolving $\text{Na}_2[\text{MoO}_4] \cdot 2\text{H}_2\text{O}$ and $[\text{MoCl}_5]$ in HCl, respectively. Another Mo(V) solution were also prepared by reducing Mo(VI) solutions electrochemically. Carrier-free radiotracer $^{181}\text{W}(T_{1/2}=121.2 \text{ d})$ was used in the extraction of W(VI). ^{181}W was produced via $^{181}\text{Ta}(p, n)$ reaction at the AVF cyclotron in RCNP of Osaka University. W was dissolved in HCl after the separation from Ta target. The aqueous phase containing Mo(V), Mo(VI) or W(VI) was mixed with equal volume of the Aliquat 336 in chloroform, and the mixture was shaken for 20-30 min at about 25 °C. Subsequently, phase separation was made by centrifuging the mixture. After the extraction, the amounts of Mo and W were determined by UV-visible spectrometry for Mo and X-ray measurement for W, and the distribution ratios were obtained. We are going to report the variation of the distribution ratios of Mo and W against the HCl and Aliquat 336 concentrations.

1P18 Excitation function for fusion reaction of a very deformed nucleus of lutetium-175

Toda, K., Kaiya, H., Yokoyama, A. (Kanazawa Univ.) , Takahashi, N. , Kasamatsu, Y. (Osaka Univ.) , Ooe, K. (JAEA)

We have studied the effect of the degree of deformation in the fusion reaction of a deformed nucleus-spherical nucleus. In this study, we measured cross sections of the fusion reaction of $\text{Lu} + \text{O}$. The Lu-175 nucleus is known to be more deformed than Tm and Tb, which were used in our previous studies. The excitation functions in the $^{175}\text{Lu}+^{16}\text{O}$ system were determined to evaluate its systematics together with the results of previous studies. The evaporation residue cross sections were measured by using a stack technique and a gas-jet technique in the projectile energy range of 100 to 150 MeV. By gamma-ray spectrometry, $^{186-188}\text{Au}$, $^{186-188}\text{Pt}$ and $^{184-187}\text{Ir}$ were detected and the cross sections were obtained from the activities. The cross sections were compared with the theoretical values calculated by ALICE code and HIVAP code. The measured excitation functions were found to exhibit a shape broader than calculation. The mechanism of interest is currently under discussion.

1P19 Reexamination of the procedure preparing for reversed-phase extraction chromatography resin for speciation of rutherfordium

Takeda, Y., Shigeyoshi, Y., Yokoyama, A., Toyoshima, A. (Kanazawa Univ., JAEA)

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. We had

used kel-F particles as substrate of TTA, the diameter of which is around 0.5 mm and not of suitable size for micro-columns (1.6 mm×7 mm). In this study, we used CHP20Y of 30 μm in diameter with a newly devised procedure of preparing for TTA resin. It was experimentally found that the TTA resin thus prepared has the extraction ability which responds to the property of liquid-liquid extraction.

1P20 Development of Gas-jet coupled Surface Ion Source of ISOL for Measurement of First Ionization Potential of Lr

Sato, T. K., Sato, N., Asai, M., Tsukada, K., Toyoshima, A., Kaneya, Y., Ichikawa, S., Nagame, Y., Schädell, M. (JAEA, Ibaraki Univ., RIKEN)

The first ionization potentials of the heavy elements with atomic number $Z > 100$ have never measured owing to the low production rates and short half-lives of the nuclides. In order to determine the first IP of the heaviest actinide element, lawrencium (Lr, $Z = 103$), by the surface ionization comparison method, we have developed a surface ionization ion source coupled with a gas-jet transport system for an ISOL (Isotope Separator On-Line) at the JAEA tandem accelerator facility. From the results of our fundamental experiments by using short-lived lanthanide isotopes, we have decided to apply some improvements to the apparatus to increase surface temperature of the ion source and reduce the cold spots, and achieve stable operation for a long beam time.

In the conference, we will mention the current status of the ion source development and the improvements.

1P21 Basic study of gas-phase chemistry using RIKEN GARIS/gas-jet system.

Kojima, T.¹, Murayama, H.¹, Murakami, M.¹, Goto, S.², Haba, H.³, Kaji, D.³, Morimoto, K.³, Kudou, Y.³, Morita, K.³, Kikunaga, H.⁴, Sato, T. K.⁵, Tsukada, K.⁵, Kudo, H.⁶ (¹Grad. School of Sci. and Technol., Niigata Univ., ²Center for Instrumental Anal., Niigata Univ., ³Nishina Center, RIKEN, ⁴Tohoku Univ., ⁵Advanced Sci. Res. Center, JAEA, ⁶Fac. of Sci., Niigata Univ.)

A gas-phase chemical separation method which examines behavior of adsorption and desorption of volatile compounds on a column surface is one of the most useful techniques to investigate chemical properties of superheavy elements. An adsorption enthalpy is usually derived by this method using Monte Carlo simulation. However, if two isomers which have different half-lives each other can be measured, it is expected that the adsorption enthalpy is derived without the parameters of experimental conditions. Recently, it was reported that ²⁶¹Rf has the isomer with 1.9-s half-life. Therefore, if the gas-phase chemistry for ²⁶¹Rf with 68-s half-life and that isomer can be performed, new analysis can be applied. The aim of this work is to perform the experiment of the gas-phase chemistry of the nuclides with short half-lives using a gas-filled recoil ion separator (GARIS) coupled with a gas-jet transport system.

As the test experiment, the gas-phase chemistry of Hf isotopes was performed. Hf isotopes were produced in ^{nat}Gd(¹⁸O, xn) reaction at the RIKEN linear accelerator facility. The reaction products separated from the beam and by-products by GARIS were introduced to the gas-jet chamber. The products were attached on a carbon cluster in

the gas-jet chamber, and were transported to the apparatus of the isothermal gas chromatography by a flow of a He carrier gas. The transported products were then collected on quartz wool plugged in a quartz tube, and formed volatile chloride compounds by a reactive gas of HCl. The volatile compounds of Hf passing through the isothermal column were attached to KCl aerosol particles, and were re-transported to the collection site for gamma-ray spectrometry. The pressure in the gas-jet chamber has to be lower than 60 kPa because the film to separate from GARIS is not so strong. On the other hand, the carbon cluster does not generate at the pressure of lower than 120 kPa. To solve this problem the carbon cluster generator and the gas-jet chamber were connected by a Teflon capillary (1.0 mm i.d. × 6, 10, 16 m length). The transport efficiency was determined as the function of the length of the capillary and the flow rate of He carrier gas.

1P22 Extraction of Zr and Hf in TIOA/HCl system for online batch extraction of Rf with the resin

A. Kino, Y. Komori, Y. Kikutani, T. Yokokita, Y. Kasamatsu, N. Takahashi, T. Yoshimura, and A. Shinohara (Osaka Univ.)

We are interested in the chloride complex formation of rutherfordium (Rf, $Z = 104$). By the past pioneering researches, basic chemical properties of Rf have been studied by the chromatographic method. In many cases, however, achievement of chemical equilibria was not confirmed. Purpose of the present study is to obtain the extraction data of Rf in equilibrium and to clarify the properties in complex formation of Rf. In this work, we performed liquid-liquid extractions and solid-liquid extractions of Zr and Hf, the homologues of Rf, in the triisooctylamine (TIOA)/HCl system by the batch method. We obtained the distribution ratios (D) and distribution coefficients (K_d) of Zr and Hf and the dependence of the values on the shaking time. Moreover, we examined the dependences of the D values and the K_d values on TIOA concentration and HCl concentration. Based on the present results, we propose suitable experimental conditions for the extraction of Rf.

1P23 Mössbauer absorption anomaly of gamma radiation via nuclear level anticrossing in FePSe₃ single crystal and powder sample

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By using FeCO₃ and FePSe₃ single-crystal absorbers, 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 order to clarify whether FePSe₃ can be an appropriate absorber for EIT experiments or not, an extensive study was carried out using powder samples of FePSe₃. A single crystal of FePSe₃ was synthesized by solid phase reaction in the vacuum-shielded tube (Vycor glass tube) including the mixture of stoichiometric amounts of Fe, P, Se at 840°C for about 1 month. The powder sample of FePSe₃ was made by grinding the single crystal thus synthesized.

From a series of Mössbauer measurements for FePSe₃ at various temperatures, we observed that at around 30K the

states of $|I=3/2, m=-3/2\rangle$ and $|I=3/2, m=1/2\rangle$ cross and are mixed, and found an obvious deficit in Mössbauer absorption at colliding peaks. 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.

1P24 Analysis of properties of hydrothermal synthesized kimzeyite by Mössbauer spectroscopy

KAMIJYO, R., SAITO, T. (Osaka Univ.), KAWASE, M. (Nagahama Inst. Bio.), YAMAKAWA, J. (Okayama Univ.)

Kimzeyite has the composition, $\text{Ca}_3(\text{Zr}, \text{Ti})_2(\text{Si}, \text{Al}, \text{Fe}^{\text{III}})_3\text{O}_{12}$, and belongs to garnet groups mineral. Garnet minerals generally have the stable structure and hardly exchange elements in them with environment. Kimzeyite also shows the long term stability of its structure like other garnet minerals, and is expected as a stabilizer in stratum disposal of the radioactive waste. It should clarify the mechanism of a long term stability to apply kimzeyite as stabilizer.

Kimzeyite samples with various Al/[Al+Fe] ratio were hydrothermal synthesized. And their structure and the state of Fe in them were analyzed by X-ray powder diffraction (XRD) and Mössbauer spectroscopy.

Mössbauer spectroscopy showed that Fe occupied the Zr site as well as original Fe site with increase of Al/[Al+Fe] ratio. XRD showed that the lattice constants of samples were not change with increase of Al/[Al+Fe] ratio. The obtained results suggest that the change of Fe occupied site relaxes a distortion of kimzeyite lattice.

1P25 Iron-57 Mössbauer spectra of iron-porphyrin complexes in frozen solution state.

Takahashi, M., Ikezaki, A., Nakamura, M. (Toho Univ.)

A Mössbauer cell for the frozen solution measurements is newly designed. The cell, made of SUS303, has a effective size of 11 mm in diameter and 5 mm in length for γ -ray transmission. A 50 μm Al foil for the window is sealed with indium wire. Iron-57 Mössbauer spectroscopy is used for the titration of $[\text{Fe}(\text{TMP}\cdot)(\text{ClO}_4)_2]$ toluene solution with NH_4F . methanol solution at 193 K. Three Fe(III) species are observed in the starting $[\text{Fe}(\text{TMP}\cdot)(\text{ClO}_4)_2]$ solution. New Fe(III) species, which is assigned to $[\text{Fe}(\text{TMP}\cdot)\text{F}(\text{MeOH})]$ or $[\text{Fe}(\text{TMP}\cdot)\text{F}(\text{ClO}_4)]$ appears after the addition of one equivalent NH_4F . The spectrum are changed finally to that of solo species assigned to $[\text{Fe}(\text{TMP}\cdot)\text{F}_2]$ after the addition of two equivalent NH_4F . The present experiments indicate clearly that the one-electron oxidation species formulated as $[\text{Fe}(\text{TMP})\text{F}_2]$ is the Fe(III) complex of porphyrin radical cation.

1P26 Hyperfine fields of perovskite manganite $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{1-x}\text{Fe}_x\text{O}_3$ ($0.2 \leq x \leq 1$) measured by Mössbauer spectroscopy

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Perovskite manganese oxides ($A\text{MnO}_3$) 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 Mössbauer spectroscopy to a study of a perovskite $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{1-x}\text{Fe}_x\text{O}_3$ ($0.2 \leq x \leq 1$). Powder $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{1-x}\text{Fe}_x\text{O}_3$ ($0.2 \leq x \leq 1$) was synthesized by a conventional solid-state reaction in air and Mössbauer measurements were performed at room temperature. Fe concentration dependence was observed in the Mössbauer spectra; while the spectra for 20% and 40% Fe-doped samples can be fitted by assuming a doublet component, coexistence of a sextet component was observed in the spectra for 60%, 80%, and 100% Fe-doped samples. These results suggest that the room-temperature magnetism of the samples changes depending on the Fe concentration.

1P27 Effect of radiation irradiation on local fields in ZnO

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Zinc Oxide (ZnO) is a material strongly expected for application as a semiconductor. When semiconductors are doped with impurities, their conductivity varies depending on the doping methods, type and density of the impurities. To control the conductivity of semiconductors, it is necessary to understand the sites and functions of the 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 ¹¹¹Cd (\leftarrow ¹¹¹In) probe. As a result, we observed spectral differences from the undoped samples: for example, the magnitude of electric field gradients (EFG) at the probe sites. In general, when impurities are introduced into materials, lattice defects are formed. In this work, we aimed to investigate if such defects have correlation with the variation of local fields in the vicinity of the probe in ZnO. For this purpose, we obtained TDPAC spectra for polycrystalline ZnO samples doped with the ¹¹¹Cd (\leftarrow ¹¹¹In) probe, which were irradiated with electrons and bremsstrahlung radiations in order to form lattice defects.

For the spectra of the irradiated samples, compared with the unirradiated, the change of the electric quadrupole frequency was not observed but the attenuation of spectral amplitude was. The attenuated amplitude, which recovered during the measurement at 1000 K, is explained as broadening of the frequency distribution. As a result, we infer that the radiation irradiation made disorder in the crystal structure causing the frequency distribution.

1P28 Observation of hyperfine fields at impurity Fe sites in ZnO

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Zinc oxide (ZnO), one of the most promising semiconductors, is expected to show room-temperature ferromagnetism by doping dilute magnetic metal. It is reported that lattice defects take an important role in showing the magnetism; however, the origin of the magnetism is still in controversy. In the present work, we prepared Fe-doped ZnO by sintering a mixture of ZnO powder and iron nitrate. The sample was irradiated with bremsstrahlung radiations and electrons, or with an ¹⁶O beam to investigate the magnetic properties before and after the irradiation by Mössbauer spectroscopy. In the Mössbauer spectra for all the samples irradiated with bremsstrahlung, a magnetic component was not observed. However, the value of QS became smaller by the irradiation of bremsstrahlung radiations for the sample doped with 2 at.% Fe. In the presentation, we discuss the Fe content dependence and the effect of the irradiations based on the Mössbauer parameters.

1P29 Molecular effects of the muon transfer process in the mixture solution of cyclohexane and carbon tetrachloride.

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In the negative muon capture on the hydrogen-containing molecule, the muonic hydrogen atoms become free from the chemical bond, and transfer the muon to other heavier atoms. The purpose of the present study is to obtain information on the molecular effect in the transfer. In this work, the measurement of muonic X-ray spectrum was performed for the mixed C₆H₆+CCl₄ or C₆H₁₂+CCl₄ solutions with various mixing ratios. Muon irradiation was performed in the D2 muon beam line of J-PARC-MUSE. Aluminum sample holder filled with a sample solution was set in the gas chamber filled with helium gas. Two germanium detectors were set on both sides of the chamber for the muonic X-ray measurement. We analyzed muonic X-ray intensity of carbon and chlorine for these samples. Obtained intensity value of each transition was normalized μ C(2-1) or μ Cl(3-2) intensity value. Here, (2-1) represents muonic transition from the state with the principle quantum number n=2 to n=1. We compared the experimental values of relative muonic X-ray intensities with those obtained by the cascade calculation to discuss the molecular effect.

3P01 Measurement of ¹⁰Be with JAEA-AMS-TONO

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The JAEA-AMS-TONO system has developed ¹⁰Be-AMS for geochronological studies to estimate exposure age of basement rocks and environmental science study. System condition for ¹⁰Be-AMS was optimized and test measurements using several standard samples and unknown samples were carried out in order to assess the accuracy of the measurements.

A gas ionization detector with a built-in gas absorber

cell was installed to prevent ¹⁰B from interfering with detection of ¹⁰Be. The gas absorber cell was composed by Havar foil and nitrogen gas in which pressure was optimized to prevent ¹⁰Be efficiently. Two standard samples with known ¹⁰Be/Be ratios supplied from the University of California, Berkeley were used to check our system performance. The results of the standard samples were almost corresponding to the certified values. Moreover ice core samples collected from the Dome Fuji station, earthen Antarctica were measured. The results were as the same level as those measured by MALT, Univ Tokyo.

3P02 Installation of Auto sample changer for the Prompt Gamma-ray Activation Analysis instrument at the JRR-3M, JAEA.

HATSUKAWA, Y., OSAWA, T., (JAEA)

Auto sample changer using the Selective Compliance Assembly Robot Arm was installed to the Prompt Gamma-ray Activation Analysis instruments at the JRR-3M. The auto sample changer has 20 sample holders. Control of the Robot arm and MCA for gamma-ray measurements, opening and shutting of upper door of measurement chamber and the neutron shielding shutter of beam line, are controlled by the LabVIEW program.

3P03 Biodistribution of Gadolinium-based contrast agent, and concentration of trace elements in normal and nephrectomized rats

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We measured distribution of Gd and concentration of some minerals in selected tissue of two type rats by using neutron activation analysis. One is normal, and the other is partially nephrectomized, which is used as pseudo renal impairment model.

The biodistribution of gadolinium 2-d after administration could be determined by ¹⁵³Gd (T_{1/2} = 240.4 d). The kidney showed high Gd retention behavior due to normal process of excretion of Gd. Skeletal Gd distribution of model rats was significantly higher than that of normal rats. All of three nephrectomized rats showed high Gd retention in each tissue compared to normal rats.

The concentration of minerals and halogens such as Na, Cl, K, Mn, Ca, Fe, Zn, and Br could be determined here. Among them, zinc had been considered to have re-levance to unchelated Gd (free Gd²⁺ ion). Concentration of zinc in femur of model rats was higher than that of normal rats.

3P04 Highly-precise radiocarbon measurements using the Compact-AMS system at Paleo Labo Co., Ltd.

Ozaki, H., Itoh, S., Niu, E., Hirota, M., Lomtadze, Z., Jorjoliani I., Yamagata, H., Kobayashi, K., Fujine, H. (Paleo Labo Co., Ltd.)

Paleo Labo Co., Ltd., installed a compact accelerator mass spectrometer (AMS) system in 2004 and some improvements have been carried out until now. Few thousand samples are measured every year and more than 15,000 samples have been measured so far.

In order to investigate the performance of our compact AMS system, we usually measure several evaluation standards distributed by IAEA. The precision of each

measurements of IAEA C6, C5 and C1 were around 0.1~0.3%, 0.3~0.5% and 2~7%, respectively and seem to be fairly good. Our procedure blank calculated from the measurements of IAEA C1 was 0.147 ± 0.034 pMC corresponding to 52,395 year BP as radiocarbon age. The average and standard deviation of reputable measurements of IAEA C6 and C5 were 150.21 ± 0.28 pMC and 23.10 ± 0.14 pMC, respectively. For C5, our result agrees with information value from IAEA. On the contrary, the result of C6 was slightly lower than the information value. However, many researchers suggest that the information value of C6 from IAEA is high. Our result of IAEA C6 agrees with the value reported by Xu et al. (2010), 150.16 ± 0.02 pMC. It's certified that our radiocarbon measurements have fairly good precision and accuracy.

In this paper, we will also present some examples of outcomes by the highly precise radiocarbon measurements performed by Paleo Labo Co., Ltd..

3P05 Elimination of ^{131}I and ^{137}Cs in rainwater from Fukushima Daiichi Accident

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Behavior of ^{131}I , ^{134}Cs , and ^{137}Cs in environment, released from the Fukushima Daiichi plant were investigated. Most of the ^{134}Cs and ^{137}Cs from rainwater were trapped by the surface soil and sand to a depth of 1 cm, whereas only about 30% of the ^{131}I was collected by the surface soil. The ^{131}I in the rainwater was in the anion exchangeable form, and all of it could be collected by anion exchangeable mechanisms, whereas 30% of the ^{131}I that had passed through the soil could not be trapped by the anion exchange resin, suggesting that the chemical form of this 30% was in a changeable, organic-bound form. The ^{131}I , ^{134}Cs , and ^{137}Cs that were absorbed on soil were difficult to be dissolved into water. As the half-life of ^{131}I is short and ^{137}Cs is strongly adsorbed on the surface soil and sand, these radionuclides would be unlikely to reach the groundwater before completely decaying; contamination of groundwater with ^{131}I and ^{137}Cs supplied from rainwater to the surface soil is therefore exceedingly unlikely.

3P06 Imaging technique for detection of leaf surface contaminations with fission fragments

Furuta E. (Ochanomizu Univ.)

Fission fragments by the Fukushima daiichi catastrophe were scattered over a large area. Between Tokyo and Fukushima, leaves were gathered up from March to June and measured their radioactivities by HPGe. Radionuclides of $^{129\text{m}}\text{Tc}$, ^{131}I , ^{134}Cs and ^{137}Cs were rightly detected. Also the leaf surface contaminations were shown as images by imaging technique which detected beta- and gamma-rays exposure. The clear images were gotten within 24 h exposure though a normal leaf image needs at least one week exposure in a shield box. The total PSL values of leaves depend on their activities of sampling points, so the imaging technique is able to use for detection of the spread of fission fragments and the relative concentration. Also the rain effect to leaves could be shown by imaging technique when leaves were washed in running water. The imaging technique is easy and useful for the nuclear catastrophe.

3P07 Observation of Atmospheric Radioactive Lead Isotopes at Kawasaki, Japan

Koike, Y.¹, Kurihara, Y.¹, Takahashi, M.², Sato, J.¹ (Meiji Univ.¹, Central Research Institute of Electric Power Industry²)

Atmospheric concentrations of radioactive lead isotopes were observed with aerosol sample intermittently during April to July 2011 at Kawasaki. Spectral lines from ^{210}Pb , ^{212}Pb , ^{214}Pb , ^{208}Tl , ^7Be , ^{131}I , ^{134}Cs and ^{137}Cs are identified in the spectrum of aerosol sample. The concentrations of ^{210}Pb and ^{212}Pb at Kawasaki ranged from 0.30 to 0.65 mBq/m³ and from 0.9 to 3.1 mBq/m³, respectively. These concentration levels are similar to a past values observed at Kawasaki. Observations of atmospheric ^{131}I , ^{134}Cs and ^{137}Cs are estimated to be responsible to the transportation of ^{131}I , ^{134}Cs and ^{137}Cs originating from the Fukushima nuclear accident.

3P08 Studies on temporal change in depth profiles of the Rn concentrations in natural water columns

Yamada, N., Maeda, E., Uesugi, M., Sato, W., Yokoyama, A., Nakanishi, T. (Kanazawa Univ.)

The Rn concentrations are measured in atmosphere and water in many places on the earth. In order to determine the radon data in water sample, special attention should be given to the radon loss from the sample. In this study, laboratory experiments by using spring water have been performed for the purpose. We have measured the water depth profile of residual Rn for water depth of 1 m with elapsed days and water temperatures. The results show that total residual Rn in water decreases exponentially in the time scale of days and the radon decreases more rapidly as the water temperature decreases. And we analyzed the results with a mass balance model with diffusion parameters. The calculation data are in good agreement with the measurements.

3P09 Leaching experiments of radionuclides from monazite

Horiuchi, K., Minowa, H., Yoshizawa, Y. (The Jikei University School of Medicine)

Many environmental radioactive nuclides are dissolved from the crust by chemical reactions with their surrounding materials. If this mechanism can be understood accurately, various applications would be expanded. In this experiment, the radionuclides were leached from a monazite using the organic acid. The 100 meshes monazite from Malaysia was used as the experimental sample. 20ml of 0.1% citric acid and 5g of the monazite were reacted at 40°C. The leached radioactivity was measured with the liquid scintillation counter LSC-6100 (Aloka). Although the existence of ^{232}Th and ^{224}Ra in the leachate was admitted, the existence of other radionuclides was not confirmed clearly. Gamma-ray spectrometry of the leachate from the same monazite has been reported the existence of ^{212}Pb and ^{212}Bi that indicates the existence of ^{220}Rn .

3P10 Analysis of Radionuclides from Fukushima Nuclear Power Plant Accident in Machida and the Surrounding Area

ENDO, K., NAKAMURA, K., TSUCHIDA, T., MATSUOKA, K., HONDA, C., TSUKADA, M. (Showa Pharm Univ.)

The radioactivity in surface soil at Kanagawa, Tokyo, Chiba and the surrounding area have been measured during the period from March to July 2011 after the accident of Fukushima Daiichi Nuclear Power Plant arising from the Tohoku earthquake and tsunami. The soil samples were dried at 97°C for 20-24h, and weighted. The γ -ray spectra were measured with a high-purity germanium semiconductor detector equipped with a multichannel analyzer. The radioactivity level of ^{137}Cs was found to be a few Bq/kg to one hundred in 15 samples with the average of 43 Bq/kg on April 1 as the compassion date, and that of ^{134}Cs to be nearly the same range (0.5-142 Bq/kg) with the average of 49 Bq/kg. The radioactivity level of ^{131}I was 20-60 Bq/kg at lower limit and several hundred Bq/kg at upper limit, and found to be higher than that of the cesium isotopes. The radioactivity ratio of $^{134}\text{Cs}/^{137}\text{Cs}$ was found to be 0.9-1.2 for all the samples, while that of $^{131}\text{I}/^{137}\text{Cs}$ was likely to be characteristic for the sampling area.

3P11 Distributions of radionuclides ^{129}I and ^{137}Cs in soils before nuclear accident

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^{129}I (half-life: 1.57×10^7 yr) and ^{137}Cs (half-life: 30.17 yr) are produced both naturally and through human nuclear activities. The majority of these nuclides will be accumulated in surface soils or ocean by global fallout. ^{129}I is important long-lived radionuclide used as estimation of ^{131}I abundance. We determined radioactive concentration of these nuclides to investigate the radioactive damage before and after nuclear accident at the Fukushima No.1 nuclear power plant. The study aims to report ^{129}I and ^{137}Cs activities in Fukushima soils before nuclear accident. Isotopic ratios of $^{129}\text{I}/\text{I}$ were determined by accelerator mass spectrometry (AMS) and ^{137}Cs concentration were measured by an HP-Ge detector. Samples were collected from 18 points at Fukushima prefecture. We report the distribution of long-lived radionuclides in surface soils (0~10 cm) at all points and 1 meter depth profile at four points. Radioactive concentration of surface soils are 0.16 ~ 0.65 (mBq/ kg) for ^{129}I and 12.60 ~ 44.40 (Bq/ kg) for ^{137}Cs . These results are standard value by comparison with previous data in Japan.

3P12 Depth profile of *in situ* cosmogenic ^{36}Cl in limestone under ground level: for the reconstruction of denudation rates

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Calcite is a main target mineral that accumulates cosmogenic ^{36}Cl (half-life: 3.01×10^5 yr). The main ^{36}Cl production mechanism near the ground surface is spallation of ^{40}Ca . While production of ^{36}Cl under the several meters depth is mainly due to cosmic ray muon. The muogenic reactions are direct negative muon capture by Ca: $^{40}\text{Ca}(\mu^-, \alpha)^{36}\text{Cl}$, and capture by ^{35}Cl of secondary neutrons produced via muon capture and muon-induced photodisintegration

reactions. Limestone is desired to be used for exposure dating and/or determine of denudation rates by cosmogenic ^{36}Cl in calcite for the study of development of karst landforms.

In the present study, limestone sample were collected from 0 to 3 m depths below surface at two outcrops in Akiyoshi karst, Yamaguchi, SW Japan. Concentration of cosmogenic ^{36}Cl was measured by AMS (Accelerator Mass Spectrometry) at University of Tsukuba. Chemical component of the limestone was quantified by prompt γ ray analysis (PGA) at Japan Atomic Energy Agency. Production rates of ^{36}Cl via ^{40}Ca spallation and neutron capture by ^{35}Cl were then calculated from the PGA results.

The ^{36}Cl activity decreases exponentially with increasing depth. Denudation rates of the land surface were calculated using the scaled ^{36}Cl production rates by an inverse analysis. The denudation seems to have increased through the Last Glacial to Holocene climate transition.

3P13 Radioactive nuclides in soil at northern Ibaraki prefecture from Fukushima Daiichi Nuclear Power Plant.

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We have measured radioactivities in soils contaminated by the severe accident of Fukushima Daiichi Nuclear Power Plant. The soil samples were taken at the 83 points distributed around the north area of Ibaraki prefecture. Radioactivities in the soils were measured by gamma-ray spectrometry, and distributions of ^{131}I and $^{134,137}\text{Cs}$ in this area were clarified. The highest radioactivity of $^{130}\text{kBq/m}^2$ of ^{137}Cs was observed at the boundary among Tochigi, Fukushima, and Ibaraki prefecture, while it was about $^3\text{kBq/m}^2$ at the center of Ibaraki.

3P14 Correction of sum-coincidence on Ge detectors using nuclide mixed activity standard volume source

Hamajima, Y. (Kanazawa Univ.)

In the gamma-ray spectrometry of the cascade gamma emitting nuclides using a Ge detector, the coincidence sum effect must be always corrected, even at low count rate. As affected by the scattered radiation, and because it depends on Ge detector, shielding material, and also measured sample, this correction must be measured in each case. Correction method has been reported in many ways. In the laboratory without RI controlled area, the reported correction methods may not be applied, because usual gamma-ray source is only commercially available for the mixed activity standard volume source. I have examined the correction method of the coincidence sum effect, using this usual standard source.

3P16 Solvent extraction of Bk and Md into HDEHP from HNO_3 solution

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Center, Japan Atomic Energy Agency.)

Whereas the chemical research on heavy actinides An(III) is difficult to be performed, it is very important to systematically study the chemical properties of An(III) and to compare these properties with those of lanthanides Ln(III) in order to discuss the properties of 4f and 5f elements. Previously, we systematically studied the extraction behaviors of Ln(III) as well as those of ^{95}Am , ^{96}Cm , ^{98}Cf , ^{99}Es , and ^{100}Fm into HDEHP. In this work, we produced ^{97}Bk and ^{101}Md by using a powerful accelerator of JAEA and investigated the extraction behaviors of Bk and Md in the HDEHP/ HNO_3 system. Distribution ratios in the extractions were determined as a function of the concentration of HDEHP. From the distribution ratios, extraction constants, K_{ex} , of Bk and Md were evaluated. Tetrad effects were clearly observed in the K_{ex} vs. atomic number plots for An(III) and Ln(III).

3P17 Determination of trace elements and change of proteins in testers of zinc deficient 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 senses and so on. Synthesis of testosterone and spermatogenesis in testes are also affected by zinc deficiency. Eight weeks old male mice of ICR strain were divided into three groups. The first is Zn-def. group fed with Zn-def. diet, the second is control group fed with control diet, and the third group is fed with control diet for a week after one week treatment with Zn-def diet. After two weeks experimental period, their testes were removed and every four testes of each group were together homogenized in a Tris buffer. Then, cytosol fractions were separated from homogenates by centrifugal separation method. SDS-PAGE and two-dimensional electrophoresis were carried out. When the position and the number of the protein spots in gel after two-dimensional electrophoresis were compared with those ones, several differences were found between three groups.

3P18 Analysis of Radio-mineral from Togo mine, Katamo, Yurihama, Tohaku, Tottori prefecture, Japan

Kawase, M., Fujiwara, Y., Mizobuchi, Y. (Nagahama Inst. Bio.) Yamakawa, J. (Okayama Univ.), Saito, T. (Osaka Univ.)

It was considered that the beta-uranophane was contained in the sample from Togo mine, Katamo, Yurihama, Tohaku, Tottori prefecture, Japan. The sample was collected from inside of the mine in 1961, and yellowish powder-like crystals in sample maintained still highly radioactive. To identify the mineral species, X-ray powder diffraction (XRD) and a gamma-ray spectrum were measured.

The XRD pattern suggested that the beta-uranophane was contained in the mine. Gamma-ray spectrum showed that the elements of the ^{238}U decay series were included in this mine. These results showed the existence of the beta-uranophane. The crystal structure of the sample should be investigated for further characterization of the beta-uranophane from this mine.

3P19 Seasonal variations of ^{226}Ra , ^{228}Ra , and ^{228}Th activities in surface water from the East China Sea

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A total of 50 surface water samples were collected in the East China Sea (ECS), and seasonal and lateral variations of ^{226}Ra , ^{228}Ra , and ^{228}Th activities were examined by applying low-background γ -spectrometry. The ^{228}Ra and ^{228}Th activities (and $^{228}\text{Ra}/^{226}\text{Ra}$ and $^{228}\text{Th}/^{228}\text{Ra}$ ratios) on the surface exhibited notable seasonal and lateral variations ($^{228}\text{Ra}/^{226}\text{Ra} = 0.2\text{-}3.4$; $^{228}\text{Th}/^{228}\text{Ra} = 0.1\text{-}0.4$). Using the ^{228}Ra activity and salinity, fractions of the Kuroshio, shelf, and Yangtze River waters on the surface of ECS are estimated. On the other hand, the $^{228}\text{Th}/^{228}\text{Ra}$ ratio on surface reflects seasonal and lateral variation of particle flux.

3P20 Investigation on Transport Behavior of River Suspended Solids in Kuzuryu River Basin using $^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs , ^7Be as tracers

Kanamori Masaki, Seiya Nagao, Shinya Ochiai, Masayoshi Yamamoto(Kanazawa Univ LLRL)

Introduction

In the material movement from continental areas to the coastal ocean, the most is transported through the river. The ratio of the material that adsorbs the particle and is transported is very high though the density in the riverine water of the suspended particle is very low. Therefore, the material movement from continental areas to the coastal ocean can be understood by doing a qualitative analysis of river suspended solids. However, the research example in Japan is absolutely few from few of the amount of the suspended solids, and a qualitative research in the catchment scales is hardly done. In this research, it was tried to examine $^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs and ^7Be as a tracers in order to evaluate the material movement in Kuzuryu river basin located in Fukui Prefecture.

Sample and method

Stream water about 120L is gathered in each of one point in Kuzuryu river main stream and Hino river that is the tributary of Kuzuryu river, and only the suspended solids were collected by using continuous centrifugal method. After Freeze-drying and homogenizing, $^{210}\text{Pb}_{\text{ex}}$, ^7Be and ^{137}Cs were measured by gamma-ray spectrometry with a germanium semiconductor detector.

Result and discussion

The correlation between flow rate and the concentration of the suspended solids was greatly different in Kuzuryu river main stream and Hino river, and difference in the responses of the suspended solids to the flow rate between Kuzuryu river and Hino river were observed to be caused by the river characteristics. Both Kuzuryu river and Hino river, the specific activity of $^{210}\text{Pb}_{\text{ex}}$ showed the increasing tendency with an increase in the concentration of the suspended solids, and the specific activity of ^{137}Cs becomes a constant value regardless of increase in the concentration of the suspended solids. As for the specific activity of ^7Be , a big difference is seen in between two rivers moreover, it has decreased in Hino river contrary to an increase of the concentration of the suspended solids

though it increased with an increase in the concentration of the suspended solids in Kuzuryu river. It was clarified that the concentration of the suspended solids and the specific activities of three nuclides changed from these in relation to the watershed environment, the supply route of water, and the feature of the outflow process etc.

3P21 Radium isotopes in high saline groundwater

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Introduction

Up to now, Ra isotopes in groundwater have been researched. In Japan, high concentration of ^{226}Ra is found in the Arima Spa (6.2Bq/kg) etc. On the other hand, high concentration of ^{226}Ra that exceeds 100Bq/kg is found from high saline groundwater in foreign countries. Digging up large depth became possible in Japan, it comes to obtain hot water of high salinity in coastal region, and it is used as a hot spring in recent years. We found comparatively high concentration of ^{226}Ra in high salinity obtained from the coastal region in Ishikawa, Toyama, and Niigata by our previous work, and have been discussing that origin. However, as there had to be a lot of uncertain points, we need to study the matter further. Then, the relation between the salinity and the concentration of ^{226}Ra is examined for the hot springs in the west of Hokkaido where a lot of high saline groundwater is situated. And we experimented in adsorption of Ra to the mineral.

Methods

The water samples were gathered from near the source. U,Ra isotopes were separated by Fe,Ba coprecipitation and were measured by α and γ spectrometry. And the dissolved ions, the ratio of isotopes and the pH4 alkalinity were measured by using the chromatograph, the mass spectrometer and the titration of 0.1M sulfuric acid. U,Ra isotopes of the digging core sample (Kamoenai Spa.) were measured.

The sorption experiment of Ra to the mineral went by batch treatment. The mineral samples(0.01g, 0.1g, 1.0g) were added to NaCl solution(0.001M, 0.01M, 0.1M, 0.5M, 1.0M 30mL), and the pH was adjusted with the hydrochloric acid to 5.0~5.5. ^{226}Ra was added, and the mineral sample was stirred for two days. The centrifuge and the filtration were done after it had stirred it and measured by γ spectrometry.

Results

As results of the dissolved ions and the ratio of isotopes, it was guessed that the water samples originated in the mixture of the rain water and the seawater or the fossil seawater. The concentration of ^{226}Ra and ^{228}Ra were 10~5097mBq/kg, 15~7700mBq/kg, and the value of highest 5097mBq/kg was obtained in the hot spring of Kamoenai. $^{228}\text{Ra}/^{226}\text{Ra}$ radioactivity ratio of this hot spring was 1.46 and that ratio of the digging core was 1.32. The salinity and the concentration of ^{226}Ra were correlated. As result of experimented in adsorption of Ra to the mineral, the coefficient of partition grew whatever minerals were used. No contradiction to the result in the field was obtained.

3P22 Estimation on transport of riverine suspended solids in Satoyama area using ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ 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 $^{210}\text{Pb}_{\text{ex}}$ by gamma spectrometry. The low $^{137}\text{Cs}/^{210}\text{Pb}_{\text{ex}}$ activity ratios are found in surface samples of abandoned paddy rice field associated with accumulation of $^{210}\text{Pb}_{\text{ex}}$ relative to ^{137}Cs . The low ratio of $^{137}\text{Cs}/^{210}\text{Pb}_{\text{ex}}$ is also found in surface soil samples of unmanaged forest because heavy rain erodes surface soil. The river bottom sediments show relatively low $^{137}\text{Cs}/^{210}\text{Pb}_{\text{ex}}$ 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 mainly controlled by surface soil erosion from the forest area and paddy field by washout of heavy rain.

3P23 Vapor emission behavior from $\text{Li}_{2+x}\text{TiO}_{3+y}$

Mizuuchi, R. (Kanazawa Univ.), Hara, M. (Toyama Univ.), Matsuyama, M. (Toyama Univ.), Oya, Y. (Shizuoka Univ.), Okuno, K. (Shizuoka Univ.)

$\text{Li}_{2+x}\text{TiO}_{3+y}$ are promised to the tritium breeding material in fusion reactor. To understand the gas evolution behavior, the thermogravimetric(TG) analysis were carried out. We prepared dampen and dried $\text{Li}_{2+x}\text{TiO}_{3+y}$. The samples were heated up, and TG measurements were performed to find out vapor emission behavior. It was observed that H_2O emitted from ambient temperature to 250 °C and CO_2 emitted at about 600 °C and more. Weight losses were observed respectively.

3A01 Observed radioactive materials in rain water after accident of the Fukushima Daiichi Nuclear Power Station

Yasuda, K., Miyamoto, Y., Magara, M., Kimura, T. (JAEA)

On March 11, 2011, Fukushima Daiichi Nuclear Power Station had been damaged by massive earthquake and huge tsunami, named as "The 2011 earthquake off the Pacific coast of Tohoku". A lot of radioactive materials were released from the power station into the environment. In order to observe radioactive materials and those fluctuations, rain water was measured by using gamma-ray spectrometer. ^{134}Cs , ^{136}Cs , ^{137}Cs , ^{131}I , ^{132}I , ^{133}I , $^{129\text{m}}\text{Te}$, ^{132}Te , ^{133}Xe , ^{140}Ba and ^{140}La were observed in the collected rain water at Tokai-mura which is located at 100-km in the south of the power station. The release of radioactive materials from the power station into the atmosphere has not been observed since April 11 as a result of measuring the fluctuation for radioactivity of ^{137}Cs that is a typical radioactive material.

3A02 Extraction of Radioactive Cesium from Tea Leaves

Yano, Y.^{1,2}, Higaki, S.², Hirota, M.³, Nomura, K.³, Kubo, M. K.¹ (¹ICU, ²RIC, Univ. of Tokyo, ³Sch. Eng. Univ. of Tokyo)

Radioactive contamination of foodstuffs attributed to the Fukushima Daiichi nuclear disaster has become a social problem. This study investigated the extraction of radioactive cesium from the contaminated leaves to the tea. The green tea was brewed twice reusing the same leaves to study the difference in extraction of cesium between the first and second brew. Moreover, the extraction of cesium was studied in correlation to brewing time. The concentration of radioactive cesium was determined with gamma spectrometry, and the concentration of caffeine was determined with absorption spectrometry. About 40% of cesium was extracted from leaves in the first brew, and about 80% was extracted in the second brew. The extraction of cesium increased over time, and it reached about 80% after 10 minutes brew. The ratio of radioactive cesium to caffeine decreased linearly over time. This study revealed that the extraction of cesium was higher for the second brew, and a rapid increase in extraction was seen as the tea was brewed for 6 minutes and more. Therefore, the first brew of green tea, which was brewed within 5 minutes, contained the least extraction of radioactive cesium from the contaminated leaves.

3A03 Depth distributions of radionuclides in soils from the Fukushima nuclear accident

Satou, Y. (Grad. School of Life and Env. Sci., Univ. of Tsukuba), Sueki, K., Kitagawa, J., Ikarashi, S., Sasa, K. (Grad. School of Pure and Appl. Sci., Univ. of Tsukuba),

We have measured depth distributions of radionuclides in characteristic soils from Fukushima No.1 nuclear plant accidents. Some researchers already reported depth deposit and profiling in soils for long half life radionuclides environmentally released from previous nuclear accidents or nuclear bomb tests. However short half-life radionuclides have not been really investigated, because they disappear relatively quickly. In this study, we collected four characteristic soils at Fukushima Pref. Soil core samples were frozen at -20 degree Celsius and sliced them into pieces 1cm layer. The separated samples were inspected by γ -ray spectrometry (^{131}I , $^{129\text{m}}\text{Te}$, ^{137}Cs , and ^{134}Cs) with an HP-Ge detector. ^{131}I , ^{137}Cs and ^{134}Cs show the same tendency with respect to density deposit. The behavior of radionuclides in soils depends on particle size distribution of soil.

3A04 Variations in radioactivity of radiocesium in river waters from Fukushima Prefecture

Nagao, S., Kanamori, M., Ochiai, S., Inoue, M., Hamashima, Y., Yamamoto, M. (Kanazawa Univ.)

The radioactive concentrations of ^{134}Cs and ^{137}Cs in river waters from Fukushima Prefecture were measured. Samplings were carried out at 4-6 stations from Abukuma, Uta, Nitta, Natsui and Same Rivers in May 20, July 12-13 and July 27 in 2011. The radioactivity of ^{137}Cs in the river waters collected in May 20 ranges from 0.23 to 4.17 Bq/L. These values are three orders higher than that of Kuji and Tone River waters before the Fukushima Accident. The percentage of particulate forms of ^{137}Cs is 86% for the upstream Abukuma sample. The activity ratio of $^{134}\text{Cs}/^{137}\text{Cs}$ is almost constant, 0.90-0.91 and agree with that of aerosol and surface soil samples in Fukushima area.

3A05 Tests of some methods to remove I-131 from

contaminated tap water

Tagami, K., Uchida, S. (Nat'l. Inst. Radiol. Sci.)

Following the Fukushima Daiichi Nuclear Power Plant accident, iodine-131 concentrations in tap water higher than 100 Bq L⁻¹ were reported by several local governments in the Kanto Plain in March 2011. To remove iodine-131 from tap water, five methods were tested in this study, that is, (1) boiling, (2) adding charcoals from oak or bamboo, (3) activated charcoals, (4) water purifiers, and (5) reverse osmosis (RO) treatments. Boiling was shown to be not effective in removing iodine-131 from tap water; indeed even higher concentrations may result from the liquid-volume reduction accompanying this process. Adding charcoals and activated charcoal treatment could not remove iodine-131, because no reduction of iodine-131 was observed in tap water samples after these treatments. Only limited effect was found with water purifiers with first several portions; no effect was expected with further water treatment. On the other hand, the RO showed high iodine-131 removal percentage of more than 95%, although the method needs about 5-10 L water to obtain 1 L of RO treated water.

3A06 Research on hydrogen availability of soil water and atmospheric water during photosynthesis by plants

Ichikawa, R., Momoshima, N., Sugihara, S. (Kyushu Univ.)

This research is aimed to clarify tritium transformation from water to hydrocarbon during photosynthesis in plants. Plants have two kinds of pathways of leaf water used in photosynthesis; soil water and atmospheric water vapor. No difference is considered at present as far as hydrogen fixation during photosynthesis depending on the water pathways. If the water pathway results in difference in amounts of hydrogen fixation of plants, evaluation of hydrogen fixation depending on the water pathway is important and necessary for tritium assessment around nuclear facilities because tritium released to atmosphere is usual and the tritium will be taken up easier to leaf through stoma than through soil water. We examined tritium availability during photosynthesis for two water pathways using D₂O as hydrogen tracer. We report the result of soil water availability.

3A07 Distribution of U-236 in the Japan Sea

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The Japan Sea is a marginal sea of western North Pacific Ocean which is enclosed by four straits. Since the Japan Sea has similar physical process with open ocean, the Japan Sea is called "Miniature ocean" and which is a appropriate analog for the oceanic research. Recently, runup of water temperature, decrease of dissolved oxygen in deep water and escalation of marine pollution have been reported in the Japan Sea. These are urgent Pan-Japan Sea issues, which may be related to global environment change in near future. To take countermeasure to these problems, it is important to investigate the detail of seawater dynamics in the Japan Sea. In our previous studies, ²³⁶U has been found in surface soil as an global fallout nuclide from nuclear test,

which suggests that ^{236}U will be a useful tracer for geochemical dynamics. In sea water, ^{236}U appears to be dissolved as $\text{UO}_2(\text{CO}_3)_3^{4-}$ after introducing to surface seawater. Thus, ^{236}U has a possibility to be a useful tracer for ocean circulation. In this study, we attempted to get the information on ^{236}U and another global fallout radionuclides, ^{137}Cs , in the Japan Sea to investigate seawater dynamics. Sea water (7 sites), suspended solid (SS) (7 sites), and sediment (5 sites) samples were collected from the Japan Sea in the summer of 2010 (KH10-02 research cruise). ^{236}U in water, SS and sediments were measured with accelerator mass spectrometry (AMS) after appropriate sample treatment and purification for those nuclides by using anion exchanging resin. ^{137}Cs in seawater was purified and collected with ammonium molybdophosphate precipitation (AMP), and measured γ -ray with Ge -semiconductor detector. We could measure ^{236}U in the small amount of sea water samples and obtained depth profile of ^{236}U . The profile of ^{236}U is nearly same as that of ^{137}Cs in water column. The detail results will be presented in our presentation.

3A08 Study of cosmic-ray intensity fluctuations around 11 ka by ^{36}Cl measurements in the Antarctic ice core

Kurosumi, K., Sasa, K., Sueki, K., Takahashi, T., Kinoshita, N., (Univ. of Tsukuba) Matsushi, Y., (Kyoto Univ.) Tosaki, Y., (AIST) Horiuchi, K., (Hiroshima Univ.) Matsuzaki, H., (Tokyo Univ.) Motoyama, H. (NIPR)

^{36}Cl is a cosmogenic nuclide (half-life:301 kyr) produced mainly by a reaction of ^{40}Ar ($p, n\alpha$) ^{36}Cl in the upper atmosphere. The cosmogenic nuclide falls on the Earth's surface at a rate depending on the nuclide production rates and hence reflecting the cosmic ray intensity. Therefore we can reconstruct fluctuations of cosmic ray intensity, by determining the past ^{36}Cl depositional flux. Such fluctuations of cosmic ray intensity may indicate paleo solar activity and/or variations in the Earth's geomagnetic field.

This paper presents the results of cosmogenic ^{36}Cl measurements during 10.54-12.00 ka in the ice core drilled at the Dome Fuji station, Antarctica ($39^\circ 42' 12''$ E, $77^\circ 19' 01''$ S, 3810 m above sea level). ^{36}Cl in the ice was measured with the Accelerator Mass Spectrometry (AMS) system on the 12UD Pelletron tandem accelerator at the University of Tsukuba. A peak of ^{36}Cl depositional flux (3.0×10^4 atoms cm^{-2} yr^{-1}) was observed around 11 ka, corresponding to the ^{10}Be enhancement found in the same ice core. The variation in ^{36}Cl flux shows similar fluctuations in ^{10}Be flux reported previously. ^{10}Be / ^{36}Cl is estimated to be nearly constant at about 10.2 during 11.21-11.37 ka for the data obtained here.

3A09 Temporal variation of Pu isotope in water columns of the equatorial Pacific Ocean

Yamada, M.¹, Zheng, J.² (¹ Hiroshima Univ., ² Natl. Inst. Radiol.Sci.)

The $^{239+240}\text{Pu}$ concentrations and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios were determined by alpha spectrometry and double-focusing SF-ICP-MS for seawater samples from the two stations (AQ-7 and AQ-13) in the equatorial Pacific Ocean. The $^{239+240}\text{Pu}$ concentration at AQ-7 was 1.8 mBq/m^3 in the surface water and increased with depth

reaching 14.4 mBq/m^3 at 600 m depth, then decreased with depth to 2.7 mBq/m^3 at the bottom layer. The $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in the water columns from the equatorial Pacific Ocean were higher than the mean global fallout ratio of 0.18. The relative percent of the inventory of close-in fallout from the Pacific Proving Grounds were 25 % in the surface layer and 55 % at the 3000 m depth. The decreasing rate of $^{239+240}\text{Pu}$ over the depth interval 0 – 1000 m were estimated to be 0.89 $\text{Bq/m}^2/\text{yr}$ based on the difference in the $^{239+240}\text{Pu}$ inventories between AQ-7 and GEOSECS-246.

3A10 Evaluation of hypoxia in Tokyo bay by chemical states of elements in the sediments

OKOSHI, T., SHOZUGAWA, K., MATSUO, M. (Graduate School of Arts and Sciences, The University of Tokyo)

Hypoxia is observed in every summer season in Tokyo bay. Though it is estimated that dredged areas in the bay are cause of hypoxia, specific relation between hypoxia and dredged area is not revealed. In order to know hypoxia for past, we attempted to evaluate redox condition by chemical state of S, Fe and Mn in the sediments. Core samples were collected at dredged area from 2009 to 2011. Concentration and chemical states of elements were measured by neutron activation analysis and X-ray absorption fine structure (XAFS). From vertical distribution and chemical state of S, it was indicated the 2009 is more reductive than 2010. On the other hand, chemical states of Fe and Mn did not vary over the core samples. Therefore, in order to trace change of chemical species about Fe and Mn, the sediments of which oxidation-reduction potential was changed were measured by XAFS. Because difference of valence was not observed from the results, we estimated that chemical form of Fe and Mn at the time when these elements were deposited was conserved.

3B01 Measurement of excitation functions of rutherfordium isotopes in $^{248}\text{Cm} + ^{18}\text{O}$ reaction

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In the chemistry experiment of Rf, ^{261}Rf (^{261a}Rf ; $T_{1/2} = 68$ s, $E_\alpha = 8.28$ MeV) has been used because it has a relatively long half-life and decays by emission of alpha particle. In recent years, it is said that there exists a short-lived isomer in ^{261}Rf which is the descendant of ^{277}Cn and ^{269}Hs . It is reported that ^{262}Rf has also isomer having similar half-life. In this work, to confirm that the nuclide with a few second half-life is truly ^{261}Rf , the measurement of excitation function of Rf isotopes which are produced by $^{248}\text{Cm} + ^{18}\text{O}$ reaction was performed.

Experiments were performed at RIKEN linear accelerator facility using a gas-filled recoil ion separator (GARIS). Rf isotopes were produced in $^{248}\text{Cm}(^{18}\text{O}, xn)$ reaction. A $^{248}\text{Cm}_2\text{O}_3$ target having a thickness of 230 $\mu\text{g cm}^{-1}$ was irradiated by ^{18}O beams of energies of 88.2, 90.2, 94.8, and 101.3 MeV at the middle of the target. The decay of produced nuclides separated from the beam and the

by-products by GARIS were measured at GARIS focal plane using Si detector box. The beam ON/OFF method was applied, that is, activities accumulated in beam ON period were measured in beam OFF period.

The alpha spectra were observed under very low background conditions. The ^{261a}Rf and the daughter nuclides, ^{257}No , were clearly observed in each beam energy. Normalizing the cross section of ^{261a}Rf as 13 nb, the constructed excitation function of ^{261a}Rf was in agreement with that reported previously. It is found that the excitation functions of spontaneous fissioning nuclides, ^{261b}Rf and ^{262}Rf , were different with each other.

3B02 Fission fragment anisotropy in heavy-ion-induced fission of actinides II

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We have measured fission fragment angular distributions in the reaction of $^{16}\text{O} + ^{238}\text{U}$ ($\alpha=0.874$) at the JAEA tandem accelerator to study the effects of entrance channel mass asymmetry $\alpha=(A_t-A_p)/(A_t+A_p)$ on fission fragment anisotropy, A_t and A_p being respective mass numbers of target and projectile nuclei. The ^{16}O beam energies of $E_{\text{lab}}=89.1\text{-}100.8$ were set to provide the same excitation energies of the compound nucleus in the reactions of $^{22}\text{Ne} + ^{232}\text{Th}$ ($\alpha=0.827$) and $^{12}\text{C} + ^{242}\text{Pu}$ ($\alpha=0.906$) in the previous work, producing the same compound nucleus ^{254}Fm . It becomes clear that the fission fragment anisotropies for $^{16}\text{O} + ^{238}\text{U}$ with $\alpha=0.874$ are similar to those for $^{12}\text{C} + ^{242}\text{Pu}$ with $\alpha=0.906$ but smaller than those for $^{22}\text{Ne} + ^{232}\text{Th}$ with $\alpha=0.827$. The Businaro-Gallone critical point of the liquid drop model suggests that the large anisotropy for $^{22}\text{Ne} + ^{232}\text{Th}$ compared with those for $^{16}\text{O} + ^{238}\text{U}$ and $^{12}\text{C} + ^{242}\text{Pu}$ obviously relates to the potential energy of the touching two nuclei compared with the fission potential.

3B03 Radiochemical Approach for Synthesis of Carboxy Metallofullerene

Takeuchi, E., Hamano, T., Akiyama, K., Kubuki, S. (Tokyo Metropolitan Univ.)

Carboxy EMFs (CxEMF) are one of the candidates for the drug deliver system, because these are easily coupled with the peptide which is conjugated with desired receptor of morbidity parts. ^{225}Ac is considered to be a promising nuclide for radio immunotherapy using EMFs. However, it is difficult to make clear their properties of water soluble ^{225}Ac EMFs. In this study, we report synthesis and properties of CxEMFs encapsulating La which is homologous element with Ac by radiochemical approach.

The crude fullerene samples containing La EMFs were irradiated by thermal neutron at JRR-4 reactor. La EMFs were separated from irradiated sample by AlCl_3 and dissolved to o-dichlorobenzene (DCB). After that, succinic acid peroxide was added. This solution was stirred and heated at 80 °C for 48 hours in Ar gas flow. Produced La CxEMFs were extracted by 0.2 M NaOH solution and injected into Sephadex G-25 size exclusion gel column with deionized water as eluent.

Production yield of La CxEMF was estimated to be about 60 %. As the result of Sephadex column separation, the almost all radio activity was observed in 12 to 18 min

fractions. These results indicate the presence of carboxy EMFs in these fractions.

3B04 Improvement of Metallofullerene separation using Lewis acid.

Hamano, T., Takeuchi, E., Akiyama, K., and Kubuki, S. (Tokyo Metropolitan University.)

Various methods to separate endohedral-metallofullerenes (EMFs) from other hollow species (C_{60} , C_{70} etc.) by using differences of redox potential between EMFs and hollow fullerenes have been reported. In a previous study, we reported that the separation method using AlCl_3 as Lewis acid was applicable for the separation of metallofullerenes encapsulating lanthanoid elements from hollow fullerenes. However, only 50% of metallofullerene was recovered in the condition. Aim of this work is to improve the separation method of metallofullerenes from hollow species. In this study, we quantitatively discuss about the separation yield for the $\text{La}@\text{C}_{82}$ from hollow fullerenes. $\text{La}@\text{C}_{82}$ was prepared by arc discharge method and irradiated by thermal neutron at JRR-4 reactor of JAEA. Separation yield of $\text{La}@\text{C}_{82}$ was determined by the comparison of the amount of $\text{La}@\text{C}_{82}$ before and after extraction. As a result, separation yield of $\text{La}@\text{C}_{82}$ was found to be almost 100% in the case TiCl_4 as Lewis acid in CS_2 solution for 10 min reaction time. This result shows that the best condition for the separation of the metallofullerenes from hollow species is $\text{TiCl}_4/\text{CS}_2$ for 10 min reaction time.

3B05 The evaluation of Analytical Capability of Cd and Hg in NMIJ CRM 8133a by Multiple Prompt Gamma Ray Analysis

Miura T., (NMIJ-AIST), Toh Y., Furutaka K., Kimura A., Koizumi M., Oshima M., Hara K., Kin T., Nakamura S., Harada H.(JAEA)

In this study, the analytical capability of Cd and Hg in plastic certified reference materials by Multiple Prompt Gamma Ray Analysis (MPGA) was evaluated. NMIJ CRM 8133a heavy metals in PP resin (Cd, Cr, Hg, Pb; High concentration) and BCR 681 PE resin were analyzed by MPGA at JRR-3 of JAEA. The relationship of between count rate of ^{113}Cd (the pair of 558.4 keV and 651.26keV) and Cd amount were examined by least square method. The correlation factor of the linear fit was 0.9993, quadratic fit was 0.9995. However, the repeatability of MPGA results were 1 to 5 %. The analytical results of Cd in BCR 681 was 21.0 mg/kg \pm 1.2 mg/kg (RSD: 5.7 %) . The analytical result of MPGA was in good agreement with the certified value (21.7 mg/kg \pm 0.7 mg/kg).

3B06 Non-destructive and Multi-elemental Analysis of Bulk Materials by Muonic X-ray

Kubo, M. K.¹, Nagatomo, T.¹, Suzuki, T.¹, Ninomiya, K.², Higemoto, W.³, Sakamoto, S.², Kawamura, N.³, Strasser, P.³, Shimomur, K.³, Miyake, Y.³, Saito, T.⁴ (ICU¹, JAEA², KEK³, Museum. Japan. History⁴)

Negative muons entering materials lose kinetic energy and are eventually captured by atoms to form muonic atoms. After atomic capture, the negative muon in the muonic atom experiences a cascade transition from higher energy muonic orbitals to lower ones with emission of muonic X-rays. The energy of the muonic X-ray is also about 200 times

greater than the corresponding electronic X-rays and also characteristic of the atom which captured the muon. The yield of muonic X-rays from the material is proportional to the atomic density of the element in the material to a first approximation. The energetic muonic X-ray emitted from a bulk sample can penetrate and escape to the surface of the sample without significant absorption. By measuring the muonic X-rays with a Ge detector, elemental analysis of the muon capture site in the sample is achievable. Depth profiling as deep as a few cm is possible by varying the incident energy of the muon beam. Thus we have been developing a muonic X-ray elemental analysis system at the J-PARC/MUSE facility in Japan. First results of application to an old Japanese gold coin that successfully gave elemental compositions at two different depths will be presented.

3B07 Nuclear and radiochemical study of production and utilization of radioactive astatine isotopes using lithium ion beams at the tandem accelerator of JAEA-Tokai

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An α radioactive nuclide ²¹¹At with a half-life of 7.2 h is a prospective candidate for utilization in targeted alpha radiotherapy. We have tried to measure excitation function of ²¹¹At in the reaction of 29-48 MeV ⁷Li^{nat}Pb at the tandem accelerator of JAEA-Tokai. The cross sections of radioactive products were determined by γ -ray spectrometry. It was found that the excitation functions of astatine isotopes were reproduced by a statistical calculation with the HIVAP code. Although the calculation predicted that ²¹¹At had large cross sections below 45 MeV compared with the other astatine isotopes, the intense γ -rays of the astatine isotopes with shorter half lives made it impossible to measure the 687 keV photopeak (I_{γ} =0.26%) of ²¹¹At. Besides, a chemical behavior of carrier-free radioactive astatine isotopes has been studied with a dry-chemistry method. Details will be shown in the presentation.

3B08 Challenge for establishing a simplified separation process of MA using novel R-BTP adsorbents-2

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In order to directly separate trivalent minor actinides (MA: ²⁴¹Am, ^{242m}Am, ²⁴³Am, ²⁴³Cm etc.) from fission products (FP) containing rare earths (RE) in high level radioactive liquid waste (HLLW), the authors have challenged for establishing a simplified MA separation process by extraction chromatography using a single column. We have attempted to apply a new type of nitrogen-donor ligands, R-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 ligands showed different properties such as adsorbability and stability by having different alkyl groups. Therefore, some novel adsorbents were prepared by

impregnating different type of R-BTP ligands (*isohexyl*-, *isoheptyl*- and *Cyheptyl*-BTP) into the porous silica/polymer composite support (SiO₂-P particles). This presentation deals with *i*) comparison with adsorption and desorption properties of Am and some FP in HNO₃ solution onto the novel-BTP adsorbents, *ii*) hydrolytic and radiolytic stabilities of the adsorbents, and *iii*) successful evaluation of single-column separation of Am from FP using the most promising R-BTP adsorbent under temperature control.

3B09 Solvent extraction of lanthanides and actinides using phosphine donor extractant

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Solvent extraction of trivalent actinide (²⁴¹Am and ²⁴⁴Cm) and lanthanide (Eu) ions using phosphine donor extractants such as Triphenylphosphine (TPP), Bis(diphenylphosphino)methane (DPPM) and Bis(diphenylphosphino)ethane (DPPE) were investigated. The aqueous phase were 0.10 moldm⁻³ sodium perchlorate solution containing trace amounts of radionuclides (²⁴¹Am or ²⁴⁴Cm) or 10⁻⁴ moldm⁻³ non-radioactive Eu. The organic phases were 1,2-dichloroethane containing various concentrations of phosphine donor extractants. Equal volumes of the aqueous phase and the organic phase were mixed in vial and were shaken 30 minutes at 298 K. In the results, all elements were extracted into organic phase from aqueous phase. Compared to the same experimental conditions, the distribution ratios were decreased in the following order, DPPM > DPPE > TPP. The separation factors between Am and Eu are 80 for DPPM, 40 for DPPE and 7 for TPP respectively. In all cases, the separation factors between Am and Cm were nearly 2.

3B10 Synthesis and properties of new pentavalent uranium complexes having two salophen-type ligands

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In the previous study, tetravalent uranium complexes in which an uranium was sandwiched by two salophen-type ligands with various substituents were synthesized. In the cyclic voltammetry, reversible redox waves of those complexes showed in the range of -0.4 - 0 V vs. Fc⁺/Fc and the potential varied systematically depending on the electron donating and accepting abilities of substituents. From the results, we supposed that this type of complex is stable to the redox reaction to give pentavalent uranium complex.

The pentavalent uranium complex was obtained as black crystals by chemically oxidizing of [U(salomphen)₂] (salomphen = N,N'-4,5-dimethyl-1,2-phenylenebis(2-hydroxybenzylideneimine)).

In the UV-visible spectrum of black crystal in dichloromethane, very large molar absorption coefficient was observed in the region < 500 nm, suggesting that existence of salomphen in the complex. In the IR spectrum of the crystal, the bands originated from salomphen and PF₆⁻ were exhibited. This suggests that the net charge of the complex is positive. In the ¹H NMR spectrum of the crystal in CD₂Cl₂ at 30 °C, the chemical shifts of signals were beyond the usually observed region in the diamagnetic complex and were significantly dependent on the temperature. These chemical shift values and temperature dependence were differed from those of

[U(salomphen)₂]. From these results, we identified the black crystals as [U(salomphen)₂]PF₆.