

English Abstracts

2AS1 : Diagnosis and therapy of hypoxic tumors using radioisotopes

Fujibayashi, Y., (BIRC, Fukui University)

Hypoxia is one of important characteristics of tumors, especially from the viewpoint of poor response to radiotherapy. In hypoxic tumors, most of redox components are in over-reduced states and it might be able to detect using redox-sensitive tracers. Among the various redox-sensitive compounds, we found radioactive copper (Cu) labeled Cu-diacetyl-bis(N⁴-methyl-thisemicarbazone) (Cu-ATSM) as a plausible candidate showing reductive cellular retention in hypoxic tissue selectively. Interestingly, reduction of Cu-ATSM in tumor cells was enzymatic, namely by cytochrome reductases. Using the selectivity of Cu-ATSM to hypoxic tumors, we successfully delivered Cu-64, a cell-toxic beta-emitting radionuclide for internal radiotherapy and found a remarkable therapeutic effect in tumor bearing hamster model. Cu-64 could be produced with an ultra-small cyclotron developed for PET nuclide production. This approach is considered to be useful for the diagnosis and therapy of hypoxic tumors in nuclear medicine.

2AS2 : Neutrino science pioneered by KamLAND

Suzuki, A., (RCNS, Graduate School of Science, Tohoku University)

KamLAND was proposed to aim at studying the reactor anti-neutrino oscillations with more than 100 km base-line, using the power plant reactors nearby Kamioka. Simultaneously it is expected to search for terrestrial antineutrinos, so-called geoneutrinos which are generated from ²³⁸U and ²³²Th decays inside the Earth. We also have the plan to detect the ⁷Be solar neutrinos, accompanying with patient efforts on reducing background events. In January 22, 2002, KamLAND started the data-taking. With the data samples of 162 and 766 ton-year exposures, KamLAND measured the flux of electron antineutrinos from distant nuclear reactors. We found fewer electron antineutrino events than expected from standard assumptions about electron antineutrino propagation at the 99.998% C.L. The observed energy spectrum disagrees with the expected shape in the absence of neutrino oscillation at 99.6 % significance and prefers the distortion expected from antineutrino oscillation effects. In the context of two-flavor neutrino oscillations with CPT invariance, all solutions to the solar neutrino problem except for the "large mixing angle" region are excluded. KamLAND has recently succeeded in detecting geoneutrinos and opened a new window into the Earth

interior.

3AS3 : Positron annihilation for characterization of solids: Advantages and limitations for the free volume studies

Shantarovich, V. P., (Semenov Institute of Chemical Physics, Russian Academy of Sciences)

Positron is a particle of antimatter, particularly an antiparticle of electron, and therefore is not able to survive for a long time being surrounded by the matter. Positron and electron annihilate, i.e. disappear together, emitting, in the most cases, of two portions of energy (two annihilation γ -rays) which are equivalent to the rest mass of electron or positron. Measurements of angular distribution of annihilation γ -rays, their momentum and also the time of positron survival in the matter (positron annihilation lifetime) provide one with the novel approach to the studies of substance properties. General information on the processes of positron annihilation, positronium formation and its transformations together with variety of experimental techniques is discussed. Finally, the approach is illustrated on example of free volume studies in polymers.

2AR1 : Structural studies of inorganic compounds by using Mössbauer spectroscopy of many nuclides

Takeda, M., (Faculty of Science, Toho University)

We have done the structure and bonding studies of inorganic compounds by using Mössbauer spectroscopy of ⁵⁷Fe, ⁶¹Ni, ¹¹⁹Sn, ¹²¹Sb, ¹²⁵Te, ¹²⁷I, ¹⁵¹Eu, ¹⁶¹Dy, ¹⁵⁵Gd, ¹⁶⁶Er, ¹⁹⁷Au and ²³⁷Np (twelve nuclides). Commercial Mössbauer sources of ⁵⁷Co/Pd, Ca^{119m}SnO₃, Ca^{121m}SnO₃, ¹²⁵I/Cu, Zn^{125m}Te, ¹⁵¹SmF₃ and ²⁴¹Am have been used for ⁵⁷Fe, ¹¹⁹Sn, ¹²¹Sb, ¹²⁵Te, ¹⁵¹Eu and ²³⁷Np Mössbauer spectroscopy, respectively. ⁶¹Ni/⁶¹Ni Mössbauer source for ⁶¹Ni Mössbauer spectroscopy was prepared by using ⁵⁸Ni(α ,p)⁶¹Ni reaction in cyclotron. By neutron irradiation in JAERI ¹²⁶Te(n, γ)^{127m}Te, ¹⁵⁴Sm(n, γ)¹⁵⁵Sm \rightarrow ¹⁵⁵Eu [β^- decay], ¹⁶⁰Gd(n, γ)¹⁶¹Gd \rightarrow ¹⁶¹Tb [β^- decay], ¹⁶⁵Ho(n, γ)¹⁶⁶Ho and ¹⁹⁶Pt(n, γ)¹⁹⁷Pt reactions have been made, and ^{127m}Te/Mg₃TeO₆, ¹⁵⁵Eu/¹⁵⁴SmPd₃, ¹⁶¹Tb/GdF₃, ¹⁶⁶Ho/Ho_{0.4}Y_{0.6}H₂ and ¹⁹⁷Pt/Pt Mössbauer sources have been prepared for ¹²⁷I, ¹⁵⁵Gd, ¹⁶¹Dy, ¹⁶⁶Er and ¹⁹⁷Au Mössbauer spectroscopy, respectively.

3AR2 : Study on an average equilibrium charge state of a super-heavy element moving in a helium gas –Contribution to a new super-heavy element discovery–

Kaji, D., (RIKEN)

A gas-filled recoil separator (GARIS) for the heaviest element research was installed at an experimental hall of the RIKEN linear accelerator (RILAC) facility. One of the most interesting applications by using the separator is the discovery of new elements whose atomic numbers are greater than 112. The most important parameter for the separator is an average equilibrium charge state q_{ave} of an ion moving in a used gas. We have systematically measured distributions of equilibrium charge state of heavy ions (^{169}Tm , ^{208}Pb , $^{193, 209}\text{Bi}$, ^{196}Po , ^{200}At , $^{203, 204}\text{Fr}$, ^{212}Ac , ^{234}Bk , ^{245}Fm , ^{254}No , and ^{255}Lr) moving in a helium gas by using the GARIS. And then, an empirical formula on the q_{ave} value of the heavy ion was derived as a function of the velocity and the atomic number of ions on the basis of the Tomas-Fermi model of the atom. The formula was found to be applicable to search for transactinide nuclides of ^{265}Hs , ^{271}Ds , ^{272}Rg , $^{277}\text{112}$, and $^{278}\text{113}$ produced by cold fusion reactions.

3AR3 : Radiochemical study of light fragment production at intermediate energies

Matsumura, H., (KEK)

Yields of ^7Be , ^{10}Be , ^{22}Na , ^{24}Na and ^{28}Mg produced from various targets in irradiations with bremsstrahlung having maximum end-point energies (E_0) up to 1200 MeV were measured by γ -ray spectrometry and AMS. The yields of heavier fragments were also measured in order to survey the relative contribution of fragmentation to spallation and/or fission in the mass yield curves. A distinct difference of the target mass-dependence of these yields has made it possible to disentangle the contributions of photospallation and photofragmentation processes. It was found that the neutron-to-proton ratios of targets strongly affect the formation of the nuclei by fragmentation as revealed in the yields of the isotopic pairs as well as of the single isotopes. Furthermore, the yields of ^7Be and ^{10}Be from some targets in alpha- (400 MeV) and neutron- ($E_0 = 500$ MeV) induced reactions were measured in order to compare them with the photonuclear-reaction yields. The target-mass dependences of the yields of ^7Be and ^{10}Be showed very similar trends among alpha-, neutron-, and photon-induced reactions. However, the effect of the incident particle was also

clearly observed in the yield ratios of ^{10}Be to ^7Be .

3BR4 : Study on luminescence phenomenon of tetravalent uranium in aqueous solution

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The luminescent transition of U^{4+} in aqueous solutions was discovered in the UV-VIS region at room and liquid nitrogen temperatures. The excitation spectrum indicates that the luminescence is arising from the deexcitation of 5f electron at $^1\text{S}_0$ level and no other emissions of U^{4+} in aqueous solutions were detected for other f-f transitions. All the luminescence peaks were assigned to the transitions from $^1\text{S}_0$ level to lower 5f levels. To estimate the luminescence lifetime, luminescence decay curves were measured using time-resolved laser-induced fluorescence spectroscopy (TRLFS). At room temperature, the decay curve indicated that the lifetime was shorter than 20 ns. On the other hand, the frozen sample of U^{4+} in aqueous solution at liquid nitrogen temperature showed the same emission spectrum as at room temperature and its lifetime was 149 ns in H_2O system and 198 ns in D_2O system. At liquid nitrogen temperature, the detection limit of U^{4+} was found to be 1×10^{-6} M by TRLFS. This limit is comparable with that by LIPAS (Laser induced photo acoustic spectroscopy).

1A01 : Charge distributions of primary and secondary fragments in 12 MeV proton-induced fission of ^{232}Th

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The charge distribution of secondary fragments in the 12-MeV proton-induced fission of ^{232}Th has been deduced from data of radiochemical and time-of-flight measurements. The charge distribution of primary fragments was obtained by correcting that of secondary fragments for neutron evaporation that was determined by the double time-of-flight technique. The results show that nuclear charge polarization at scission depends on fragment mass. Such nuclear charge polarization is discussed in terms of nuclear structures of fission fragments.

1A02 : Investigation of decay property of $^{229\text{m}}\text{Th}$ by photon measurement

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A Low-lying isomer of ²²⁹Th, ^{229m}Th, is expected to have an intriguing decay property because of its extremely low excitation energy. It could be a good subject of the investigation of an electron bridge (EB) mechanism. The decay process of ^{229m}Th is indicated to vary depending on its chemical state through the EB process. We have tried to detect visible and ultraviolet photons emitted from ^{229m}Th by a low-noise photomultiplier. Th-229 samples were obtained as a decay product of ²³³U and prepared as HCl solution, HNO₃ solution and so on. Th-229 was rapidly purified by chemical separation for the photon detection. In the present work, we report the results of photon measurements for each sample and discuss on the half-life of ^{229m}Th based on the previously obtained information.

1A03 : Half-life measurement for ^{229m}Th from α -decay of ²³³U

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The nuclide of ²²⁹Th has gathered much attention for years because of an excited state (^{229m}Th) with an ultra-low energy of several electron-volts. Half-life of the state can vary with its chemical state, because the transition probability from it depends on the energy state of the outer-shell electrons. Hence, the isomer is also interesting from a chemical viewpoint. We attempted to produce the extremely low energy isomer of ²²⁹Th (^{229m}Th) from alpha-decay of ²³³U for the purpose of determination of its half-life. The products above were isolated from ²³³U by using several chemical separation methods and identified by alpha-spectrometry. One can expect to detect the alpha-particles from ^{229m}Th (4.93 MeV) based on a theoretical prediction if it has a long half-life. However, such events in the expected region were not observed in the measured alpha-spectra. From the result, we conclude that half-life of the isomer is shorter than 1 hour.

1A04 : Development of online solvent extraction system with microchip

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We have investigated various methods of chemical reactions and measurements in order to research the behavior of heavy actinide elements. In this study, we employed a microchip to enable rapid solvent extraction. Microchip is a reactor vessel made of glass or plastics with etched channels of dozens to hundreds micrometers in width. Because molecules diffuse far more rapidly in such a small vessel than in an ordinary vessel of a cm size, equilibrium can be very rapidly attained in the microchip. We have confirmed that the equilibrium is achieved within one second for the extraction of lanthanide radiotracer from acetate buffer to toluene using TTA (thenoyltrifluoroacetone) as a ligand. We report the results of online experiments at RIKEN Ring Cyclotron using the microchip system.

1A05 : Formation of Anionic Chloride Complex of Rutherfordium (Rf)

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We have investigated anion-exchange behavior of element 104, rutherfordium (Rf), in HCl and CH₃OH mixed solution together with its lighter homologues, Zr and Hf. The adsorption ratios of Rf, Zr and Hf similarly increase with an increase of CH₃OH concentration. On the other hand, the chemical species of Zr and Hf were determined with an EXAFS method to be [MCl₆]²⁻ (M=Zr, Hf) in the presence of CH₃OH. These results indicate that structure of the adsorbed chloride complex of Rf is expected to be [RfCl₆]²⁻.

1A06 : Reversed-phase extraction chromatography of rutherfordium in the TBP-HCl system

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Adsorption behaviors of rutherfordium (Rf), Zr and Hf on TBP-reversed-phase resin in 7.2, 7.5, 7.8, and 8.0 M HCl solutions were simultaneously studied using a rapid chemistry apparatus. It was clearly found that the order in adsorption probability on the TBP resin for the three elements is $Zr > Hf \geq Rf$, which is different from that on the anion-exchange resin: $Rf > Zr > Hf$. In the symposium, we will discuss the chemical equilibria for Rf in the TBP system as well as the anion-exchange one.

1A07 : Fluoride complexation of rutherfordium

Toyoshima, A.,^a Haba, H.,^b Tsukada, K.,^a Asai, M.,^a Akiyama, K.,^c Goto, S.,^d Ishii, Y.,^a Sato, W.,^e Matsuo, K.,^e Saika, D.,^e Kitamoto, Y.,^e Yokoyama, A.,^f Sakama, M.,^g Oura, Y.,^h Nishinaka, I.,^a Sato, T.,^a Ichikawa, T.,^a Hirata, M.,^a Ichikawa, S.,^a Sueki, K.,^c Shinohara, A.,^e Kudo, H.,^d Nagame, Y.,^a Nakahara, H.,^h Schädel, M.,^j (^aAdvanced Sci. Res. Center, JAERI, ^bCyclotron Center, RIKEN, ^cGraduate School of Pure and Applied Sci., Univ. of Tsukuba, ^dFaculty of Sci., Niigata Univ., ^eGraduate School of Sci., Osaka Univ., ^fGraduate School Nat. Sci. Tech., Kanazawa Univ., ^gRadiologic Sci. and Engineering, Univ. of Tokushima, ^hGraduate School of Sci., Tokyo Metropolitan Univ., ^jGSI)

We present recent anion-exchange studies on the heaviest group-4 homologue, rutherfordium (Rf), in the HF and mixed HF/HNO₃ solutions. It was found that the adsorption probabilities of Rf in the HF solutions are much smaller than those of Zr and Hf and decrease with an increase of HF concentration. The variations of the adsorption behaviors of Rf, Zr and Hf in the HF/HNO₃ solutions indicate that the formation of the hexafluoro complex of Rf, $[RfF_6]^{2-}$, is remarkably weak compared to that of the lighter homologues, $[ZrF_6]^{2-}$ and $[HfF_6]^{2-}$.

1A08 : Alpha-gamma spectroscopy of ²⁶¹Rf

Asai, M., Tsukada, K., Sakama, M.,^a Haba, H.,^b Ishii, T., Nagame, Y., Nishinaka, I., Akiyama, K., Toyoshima, A., Ichikawa, T., Ichikawa, S., Sueki, K.,^c Shibata, M.,^d Kojima, Y.,^e Oura, Y.,^f (JAERI, ^aUniv. of Tokushima, ^bRIKEN, ^cUniv. of Tsukuba, ^dNagoya Univ., ^eHiroshima Univ., ^fTokyo Metropolitan Univ.)

Alpha-gamma coincidence spectroscopy for the alpha decay of ²⁶¹Rf has been performed using a rotating wheel alpha-gamma detection system coupled with a gas-jet transport system. L X rays following the alpha decay of ²⁶¹Rf were clearly observed, while K X rays and intense gamma transitions were not detected. This indicates that the gamma transitions following the alpha decay of ²⁶¹Rf should be low-energy M1 and/or E2 transitions, and that the $3/2^+[622]$ and $11/2^-[725]$ assignments are excluded for the ground-state configuration of ²⁶¹Rf.

1A09 : Identification of New Neutron-Rich Eu Isotopes

Sato, T. K.,^a Osa, A.,^a Asai, M.,^a Tsukada, K.,^a Hayashi, H.,^b Kojima, Y.,^c Shibata, Y.,^d Ichikawa, S.,^a (JAERI, ^bGrad. School Eng., Nagoya Univ., ^cGrad. School Eng., Hiroshima Univ. ^dRI center, Nagoya Univ.)

Three new neutron-rich rare-earth isotopes ¹⁶³Eu, ¹⁶⁴Eu, and ¹⁶⁵Eu produced in the proton-induced fission of ²³⁸U were identified using the JAERI on-line isotope separator equipped with a surface ionization type integrated-uranium-target-ion source. The half-lives were determined to be 5.9 ± 0.3 s for ¹⁶³Eu, 4.1 ± 0.1 s for ¹⁶⁴Eu, and 2.2 ± 0.1 s for ¹⁶⁵Eu. The assignment of these isotopes was based upon the observation of Gd K x rays in the β -coincident x/γ -ray spectra measured for the separated mass fraction. The first observation of excited state of the daughter nucleus ¹⁶⁴Gd through the β decay of ¹⁶⁴Eu was achieved. In the present work, the on-line isotope separation combined with the recently developed ion-source has proved advantageous for the identification of neutron-rich heavy Eu isotope. This technique is expected to be applied to identify the excited state of doubly midshell nucleus ¹⁶⁶Gd through the β decay of ¹⁶⁶Eu.

1A10 : Synthesis of element 113 by the reaction of ²⁰⁹Bi(⁷⁰Zn,n)²⁷⁸113

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A.,^a Zhao, Y.-L.,^h (^aRIKEN, ^bSaitama Univ., ^cNiigata Univ., ^dUniv. of Tokyo, ^eJAERI, ^fUniv. of Tsukuba, ^gTMP, ^hIHEP)

A gas-filled recoil separator (GARIS) for the heaviest element research was installed at an experimental hall of the RIKEN linear accelerator (RILAC) facility. One of the most interesting applications by using the recoil separator is the discovery of new elements whose atomic numbers are greater than 112. We performed an experiment to study a production and decay of a nuclide ²⁷⁸113 produced by the reaction of ²⁰⁹Bi(⁷⁰Zn,n)²⁷⁸113.

1A11 : Status of the ISOL-based radioactive ion beam facility at JAERI, Tokai

Ichikawa, S., (Department of Materials Science, Japan Atomic Energy Research Institute)

Construction of the radioactive nuclear beam (RNB) facility, TRIAC (Tokai Radioactive Ion accelerator Complex), consisting of the Charge-Breeding Electron Cyclotron Resonance Ion-Source (CB-ECRIS), the Split-Coaxial Radio-Frequency Quadrupole (SCRFAQ) and Interdigital-H (IH) linacs was completed in the end of July, 2004. An Isotope-Separator On-Line (JAERI-ISOL) was connected to the CB-ECRIS through a low-energy beam transport line. The TRIAC machine tuning started in August 2004, together with RNB production using a uranium-target and a 30 MeV proton beam. In February 2005, the linacs accelerated ¹⁴N²⁺ and ¹⁶O²⁺ ions with output energy of 1.1 MeV/u. Also, in March 2005, ¹³⁸Xe ($T_{1/2}=14.08$ min.) was successfully accelerated through the CB-ECRIS to the end of the linacs. The charge state of ¹³⁸Xe was bred to 20⁺ by the CB-ECRIS and acceleration energy of 1.1 MeV/u was achieved. The TRIAC will be available for users from October, 2005. In this conference, we will report the status of the TRIAC facility as well as the future plan of this facility.

1A12 : Development of FEBIAD-type integrated-Uranium-target-ion source system

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According to the JAERI-KEK joint radioactive nuclear beam (RNB) project, we have developed a FEBIAD-type integrated-Uranium-target-ion source system to produce heavy neutron-rich RNBs with particle-induced fission of ²³⁸U. In order to evaluate RNB intensities with this ion source, yields and release times of mass-separated fission products were examined. A

uranium carbide target was prepared at a uranium density of 800-mg/cm²U. The target was bombarded with a 33-MeV proton beam with intensity of about 300 nA. For the separation yield measurements of fission products, the mass separated products were collected on an aluminized Mylar tape in a tape transport system and were periodically transported to a measuring position where an HPGe detector was placed for γ -ray measurements. Separation yields of 15 elements and 82 nuclei were derived from γ -ray measurements. Most of RNBs were "cocktail" beams. The separation yield of doubly-magic nucleus ¹³²Sn was 1x10⁴ pps. However, this fraction contained almost the same amount of ¹³²Sb. Release times of Kr, In, Sn and Xe atoms were also measured.

1A13 : The interaction between trivalent 4f and 5f block elements and thiocyanate ion

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The formation constants of thiocyanate complexes of ^{152,154}Eu(III) and ²⁴¹Am(III) in trace concentrations were investigated in mixed solvent (CH₃OH + H₂O) solutions of different ionic strength. The formation constant was obtained by a back-extraction technique. The obtained results showed that the formation constant of thiocyanate complex of Am(III) was considerably larger than that of Eu(III) in 1.0 M(H, Na)(SCN, ClO₄) mixed solvent solution of methanol molefraction 0.40 ($X_{MeOH} = 0.40$). Furthermore, in paper electrophoresis, the moving velocities of the species of Eu(III) and Am(III) were investigated in 1.1 M(H, Na)(SCN, ClO₄) mixed solvent solutions. The obtained results showed that the velocity of Eu(III) were considerably larger than that of Am(III) in 0.2 ~ 0.6 M [SCN⁻] of the mixed solvent solution ($X_{MeOH} = 0.23$). The difference between the velocities of Eu(III) and Am(III) is explained on the basis of the difference of the mean charges of the species of Eu(III) and Am(III) in the solution.

1A14 : Separation of americium and curium by use of tertiary pyridine resin in nitric acid / methanol mixed solvent system

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The separation behaviors of americium and curium by chromatography using the tertiary pyridine resin with the nitric acid

/ methanol mixed solution were studied. We confirmed that the distribution coefficient depends on the concentration of nitric acid and the maximal value was obtained using 10 M HNO₃. However, the separation factor and resolution were not affected by nitric acid concentration. The influence of methanol ratio on the adsorption and the separation were also investigated. It was confirmed that the distribution coefficients, the separation factor, and the resolution increase with the methanol ratio.

1A15 : Redox behavior of uranium ion in room-temperature ionic-liquid

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Photoreduction of UO₂²⁺ by ethanol using mercury lamp was investigated in two kinds of room-temperature ionic-liquids, which consist of diethylmethyl(2-methoxy)ammonium cation (DEMMA⁺) and bis-(trifluoromethanesulfonyl)imide anion (TFSI⁻), or 1-butyl-3-methyl imidazolium cation (BMI⁺) and TFSI⁻. In the presence of 0.1 M HClO₄ and 0.1 M ethanol, the reduction of UO₂²⁺ to U⁴⁺ was observed in DEMMA-TFSI, while no reduction was observed in BMI-TFSI, suggesting the quenching of excited UO₂²⁺ by BMI⁺. Cyclic voltammogram of U⁴⁺ in DEMMA-TFSI was also measured in a three electrode system (working electrode, GC; counter electrode, Pt; reference electrode, Ag/AgCl) at scan rate range from 5 to 400 mV/s. It was found that the redox reaction for uranium ion in DEMMA-TFSI was affected sensitively by the water content in DEMMA-TFSI, and two oxidation waves and two reduction waves were identified with the oxidation-reduction pairs of uranium ion, i.e., U(IV)/U(VI) and U(IV)/U(III), respectively.

1B01: Spatial distributions of ¹³⁷Cs and plutonium in the Pacific: Results of BEAGLE2003 cruise

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¹³⁷Cs and plutonium in the ocean, which are mainly originating from the large-scale atmospheric nuclear testing, have been measured during the past five decades. Levels and their change of ¹³⁷Cs and plutonium are important not only as aspect of assessment of radiological effect but also as better understanding of oceanic processes. In 2003/4, BEAGLE2003 expedition, around the globe along about 30°S latitude, was conducted using R/V "MIRAI" by JAMSTEC. We have collected about 800 water samples to

determine anthropogenic radionuclides. Therefore we can depict the first detail distributions of ¹³⁷Cs and plutonium in the South Pacific. ¹³⁷Cs concentrations in the South Pacific surface waters ranged from 0.1 to 1.5 Bq m⁻³. The ¹³⁷Cs concentrations in the central subtropical South Pacific are the same as that in the subtropical North Pacific, whereas extremely low ¹³⁷Cs concentrations were observed at stations near the South America. ^{239,240}Pu concentrations in the South Pacific surface waters ranged from 0.5 to 4.1 mBq m⁻³, which are the same or slightly lower than that in the subtropical North Pacific.

1B02 : ^{239,240}Pu and ²⁴⁰Pu/²³⁹Pu atom ratios in the surface waters of the western North Pacific Ocean and its adjacent seas

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Surface seawater samples were collected along the track of the R/V Hakuho-Maru KH-96-5 cruise. ²³⁹⁺²⁴⁰Pu activities and ²⁴⁰Pu/²³⁹Pu atom ratios were determined for the surface waters in the western North Pacific Ocean, the Sulu and Indonesian Seas, and the South China Sea. ²³⁹⁺²⁴⁰Pu activities in surface water showed notable variation with the sample locations. The Sulu and Indonesian Seas had an average ²³⁹⁺²⁴⁰Pu activity of 3.16 ± 0.32 mBq m⁻³, which was the highest activity in this study. ²³⁹⁺²⁴⁰Pu activities of the western North Pacific Ocean were comparable with the value estimated from the GLOMARD. The lowest was from the Andaman Sea. The ²⁴⁰Pu/²³⁹Pu atom ratios were significantly higher than the global stratospheric fallout ratio of 0.18. The higher ²⁴⁰Pu/²³⁹Pu atom ratios could be attributed to local fallout Pu delivered from the Marshall Islands.

1B03: Determination of trace elements and Pu isotopes in squid liver samples by ICP-MS

Oikawa, S., Matsuda, H., Isogai, K., (Div. of Radiochem. Anal., Japan Chemical Analysis Center)

It is well known that a squid concentrate trace elements in seawater as it grows. The main purpose of this study is verification of the usefulness of ICP-MS for measurement of trace elements in squid liver and an application to determine the concentration factors. With the aim for an analytical validation of the ICP-MS method by INAA, squid sample were collected from Japanese inshore during 1981 to 2004. Concentration of 13 elements (V, Mn, Fe, Co, Cu, Zn, Rb, Sr, Ag, Cd, Cs, Th and U) was

determined by ICP-MS after ashing and digestion processes. The analytical results of nine elements were agreed well with INAA within 10%. Plutonium isotopes were also determined by high resolution ICP-MS after chemical separation. Concentration factor for Co, Zn, Ag and Cd were 10^5 to 10^6 , and Pu was about 10^3 . The analysis of trace elements in squid liver was found to be useful to study heavy metal and radioactive contamination in marine environment

1B04: Studies on the $^{240}\text{Pu}/^{239}\text{Pu}$ ratios in soil.

Muramatsu, Y.,^a Yoshida, S.,^b Yoshikawa, S.,^c Yamazaki, H.,^d Yamazaki, S.,^e (^aGakushuin University, ^bNational Institute of Radiological Sciences, ^cOsaka City University, ^dKinki University, ^eTokyo Nuclear Service).

Concentrations of Pu and the $^{240}\text{Pu}/^{239}\text{Pu}$ ratios in soil samples were determined by high resolution ICP-MS, in order to understand the origin of Pu and its behavior in the soil environment. The $^{240}\text{Pu}/^{239}\text{Pu}$ ratios in soils increased with depth, while the Pu concentrations decreased. It was also found that the $^{240}\text{Pu}/^{239}\text{Pu}$ ratios in extracts with diluted nitric acid showed a lower ratio than its bulk value. These results suggested that there should be at least two Pu fractions, i.e. fraction of a low $^{240}\text{Pu}/^{239}\text{Pu}$ ratio fixed in the top layer and that of a high ratio moved into the deeper layer. We also analyzed samples from Nishiyama area (Nagasaki), near the hypocenter of Pu bomb. Very low $^{240}\text{Pu}/^{239}\text{Pu}$ ratios of about 0.03 were found in soil samples of this area. In a sediment core sample (327cm) collected from the Nishiyama-lake, the highest Pu concentration (32Bq/kg) with the lowest ratio (0.0288) was found at the depth of around 320 cm, indicating the layer contaminated with the bomb fallout.

1B05: Time efficiency of tritium measurement in the environmental water by electrolysis enrichment 3

Sakuma, Y., Yamanishi, H., (National Institute for Fusion Science), Iida, T., (Graduate School of Engineering, Nagoya Univ.), Ogata, Y., (School of Health Science, Nagoya Univ.), Tsuji, N., (Japan Air-conditioning Service Co & Ltd.), Kakiuchi, M., (Fac. of Science, Gakusyuin Univ.), Satake, H., (Fac. of Science, Toyama Univ.), Torikai, Y., (Hydrogen Isotope Research Center, Toyama Univ.)

We have developed a new method which is able to measure more efficiently and more accurately than the conventional method. Using this new method, several measurements have been carried

out, collecting samples from 7 sampling points every month or every three month, for two years. The results were as follows; the concentrations of tap water, rain water and spring water samples were from 0.1Bq/kg to 0.6 Bq/kg and the concentrations of water vapor samples in the air at Toki were from 0.5Bq/kg to 1.0Bq/kg. We are developing more efficient and accurate method which will be able to measure tritium concentration of low level samples in about one day.

1B06 : Determination of U isotopes in marine environmental samples using ICP-MS

Zheng, J., Yamada, M., Aono, T., Kusakabe, M., (Nakaminato Lab. Marine Radioecology, Nat. Inst. Radiol. Sci.)

The determination of uranium is important for environmental radioactivity monitoring, which investigates the releases of uranium from nuclear facilities and of naturally occurring radioactive materials by the coal, oil, natural gas, mineral, ore refining and phosphate fertilizer industries, and it is also important for studies on the biogeochemical behavior of uranium in the environment. In this paper, we describe our recent work on the investigation on U isotopic compositions ($^{235}\text{U}/^{238}\text{U}$ atom ratio and $^{234}\text{U}/^{238}\text{U}$ activity ratio) in marine environmental samples, such as seawater and sediments using quadrupole ICP-MS and sector-field ICP-MS. We found that, with natural U, a precision lower than 0.5% RSD for $^{235}\text{U}/^{238}\text{U}$ atom ratio and lower than 2.0 % RSD for $^{234}\text{U}/^{238}\text{U}$ activity ratio was obtained with less than 90 ng uranium using ICP-QMS. Direct analysis of seawater for U concentration and $^{235}\text{U}/^{238}\text{U}$ isotope ratio was achieved with Element 2 SF-ICP-MS. In seawater collected from the offshore of Aomori (Pacific), the U concentration showed a good correlation with the salinity.

1B07 : U-234/U-238 disequilibrium in gypsum

Arai, K., Sasaki, K., (Graduate School of Science, Rikkyo University)

Gypsum products made from the residue of phosphate fertilizer production contain measurable amount of uranium. In this paper, we report the enrichment of U-234 by alpha-recoil in less-soluble particle contents such as silica. The sample gypsum was ground, dried, and then leached by water, 0.1 M HCl, 1 M HCl and 8 M HCl in turn. The residue was digested and leached with the mixture of hydrofluoric acid, nitric acid and perchloric acid in a pressurized PTFE container. It was found that the U-234 was enriched in the surface layer of the less-soluble particles and in equilibrium with

U-238 in the core portion.

1B08 : Etching behaviors of polycarbonate fission track detector containing uranium particles

Lee, C. G., Iguchi, K., Magara, M., Sakurai, S., Watanabe, K., Usuda, S., (JAERI)

The etching rates of fission track detectors made of polycarbonate containing uranium particles were measured after thermal neutron irradiation with fluence of 8×10^{14} n/cm², in order to study the influence of uranium enrichment on the etching rate that was calculated from the weight loss by etching. There is a strong correlation between the etching rate of detector and the enrichment E of uranium particle: the former increases as the latter increases. Particularly, the etching rate per particle was proportional to $E^{2/3}$ rather than E^1 , which is probably due to the overlapping of fission tracks. The etching behaviors of detector revealed that the existence of two different etching rate regions, a nonlinear region in the beginning of etching process and a subsequent constant region, which was explained as the opening of fission tracks and the broadening of opened tracks, respectively. The correlation between the etching rate of detector and the enrichment of uranium particle suggests that the screening of uranium particles according to their enrichment is possible by controlling the etching time.

1B09 : $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters in Japan

Nakano-Ohta, T., Sato, J., (Meiji Univ.)

The activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ in river water with short residence time is similar to that in underground water and is supposed to be dependent on the activity ratio of Th/U in the rock with which the underground water is in contact. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in river waters from Fuji volcano and from sedimentary rock area of Japan were observed. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters for Fuji volcano was smaller than that for sedimentary rock area, suggesting that the Th/U activity ratio in rock was reflected in the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river water.

1B10 : Volatilization of Po by microorganism

Momoshima, N.,^a Fukuda, A.,^b Ishida, A.,^a Yoshinaga, C.,^a (^aFac. Sci. Kumamoto Univ., ^bGrad. School Sci. Tech. Kumamoto Univ.)

Volatilization of Po by three microorganisms were examined;

Escherichia coli K-12, *Bacillus subtilis* and *Chromobacterium violaceum*. The microorganisms were pre-cultured in LB medium at 30 °C and a small portion of the pre-cultured was transferred to a culture bottle in which LB medium and ^{208}Po tracer were contained. The culture was done at 30 °C with shaking the culture bottle. The Po volatilized was collected into the trap vials in which scintillator for liquid scintillation counting (LSC) was contained. The Po activity was measured by LSC. All of the microorganisms examined volatilized Po but their ability was quite different each other. Highest ability was observed on *Chromobacterium violaceum* and then *Escherichia coli* K-12 followed by *Bacillus subtilis*, the relative magnitude of the ability was 10², 10, 1, respectively. *Chromobacterium violaceum* and *Escherichia coli* K-12 showed high volatility for the first 24 h but *Escherichia coli* K-12 showed a decrease thereafter. However high volatility was continued on *Chromobacterium violaceum* during the culture. The low culture temperature suppressed Po volatility, supporting biologically mediated Po.

1B11 : Measurement of short-lived cosmogenic nuclides in rain and their application to hydrological tracers

Kuwahara, Y.,^a Abe, T.,^a Komura, K.,^b (^aGrad. School of Nat. Sci. and Tech., Kanazawa Univ., ^bLLRL, Kanazawa Univ.)

We have succeeded to detect short-lived cosmogenic radionuclides ^{18}F (109.7m), ^{24}Na (half-life 14.96 h), ^{28}Mg (20.9 h), ^{38}S (2.83 h), ^{38}Cl (37.2 m) and ^{39}Cl (56 m) in rain using 50-60L of rainwater collected through downpour from the roof of our laboratory. Cosmogenic radionuclides were separated within 30 min by using anion and cation type resin (Powdex-PAO and Powdex-PCH) and measured by ultra-low background Ge detector in Ogoya Underground Laboratory. Fallout levels of ^7Be , ^{24}Na and ^{28}Mg were evaluated to be 0.075-3.7 Bq l⁻¹, 0.16-6.5×10⁻³ Bq l⁻¹ and 0.16-6.3×10⁻³ Bq l⁻¹, respectively.

1B12 : New evaluation method of Atomic-Bomb neutrons using $^{108\text{m}}\text{Ag}$

Komura, K., (LLRL, K-INET, Kanazawa Univ.)

A new method was proposed for the evaluation of Atomic Bomb neutrons using $^{108\text{m}}\text{Ag}$ with half-life of 418 y. This method is estimated to be 3 orders of magnitude more sensitive compared with that by ^{152}Eu if silver items such as medals coins are available. This was confirmed by the measurement of silver medal exposed to

Hiroshima Atomic Bomb at 450 m from the hypocenter. Three prominent gamma rays from ^{108m}Ag were detected by non-destructive gamma spectrometry. ^{108m}Ag was also detected in the finger ring made of brass from the victim of Atomic Bomb, hand guard of sword, brass spoon and pocket watch. This method is much more favorable than ^{152}Eu method because (1) ^{108m}Ag can be measured nondestructively without chemical separation, (2) there is no interfering gamma-rays as in the case of ^{152}Eu and (3) abundance of 3 gamma rays from ^{108m}Ag (270 %) is order of magnitude higher than that of 344 keV gamma ray of ^{152}Eu (26.7%). Furthermore important fact is that direct neutron dose can be evaluated by ^{108m}Ag if finger ring or rosary of the victim are available.

1B13 : Measurement of technetium soil-to-plant transfer factors near the Chernobyl site

Tagami, K., Uchida, S., (Office of Biospheric Assessment for Waste Disposal, National Institute of Radiological Sciences)

Technetium-99 is a pure beta emitter and its radiological half-life is 211000 y. This nuclide is one of the most important radionuclides for dose assessment because it is thought to be high mobile in soil and available to plants due to its most stable chemical form under natural surface water conditions is pertechnetate. The chemical form of Tc, however, changes with environmental conditions, so that it is necessary to obtain transfer parameters, such as soil-to-plant transfer factors (TFs), under natural conditions. Thus, in this study, we determined the concentrations of ^{99}Tc in 27 plant leaves collected near the Chernobyl site to investigate the transfer of the nuclide in soil-plant systems under natural conditions. TFs ranged from <0.016 to 0.28 for Fern, 0.009 to 0.47 for Herb and <0.006 to 0.05 for Tree groups based on the ^{99}Tc contents of the organic layers.

1B14 : Technetium(IV) oxide colloids and the precursor produced by bremsstrahlung irradiation of aqueous pertechnetate solution

Zakir, M., Sekine, T., Takayama, T., Kudo, H., (Grad. School Sci., Tohoku Univ.)

We studied the radiolytic formation mechanisms of $^{99}\text{TcO}_2 \cdot n\text{H}_2\text{O}$ colloids by bremsstrahlung irradiation of an aqueous pertechnetate ($\text{Tc}^{\text{VII}}\text{O}_4^-$) solution under different conditions; (i) pH of the target solution, (ii) initial concentration of $\text{Tc}^{\text{VII}}\text{O}_4^-$, (iii) presence or absence of radical scavengers and of seed particles of $\text{TcO}_2 \cdot n\text{H}_2\text{O}$.

Colloid particles (30 – 130 nm) of $\text{TcO}_2 \cdot n\text{H}_2\text{O}$ — a coagulate of nanoparticles with the size of 2 nm — were radiolytically produced at pH higher than 3, whereas a soluble Tc(IV) species but colloid was formed at lower pH. The soluble Tc(IV) species transformed to the $\text{TcO}_2 \cdot n\text{H}_2\text{O}$ colloid when the pH of the solution was raised to 3 or higher after the irradiation. This fact suggests that the soluble Tc(IV) species is a precursor of the $\text{TcO}_2 \cdot n\text{H}_2\text{O}$ colloid. The presence of $\text{TcO}_2 \cdot n\text{H}_2\text{O}$ seed particles in the target solution of $\text{Tc}^{\text{VII}}\text{O}_4^-$ did not enlarge of the particle size, revealing that the solidification would take place rather in the solution than on the surface of nanoparticles. The reduction of TcO_4^- in acidic solutions proceeded mainly through processes involving a bimolecular reaction of TcO_4^- with hydrogen radicals, followed by the successive disproportionation reactions of Tc(VI) and Tc(V).

1B15 : Radiolytic formation of technetium-acetylacetonate

Zakir, M., Taniguchi, H., Takayama, T., Sekine, T., Kudo, H., (Grad. School Sci., Tohoku Univ.)

The pertechnetate solution containing acetylacetonate was irradiated with bremsstrahlung generated from an electron linear accelerator (LINAC) of Tohoku University. Products in the solution were analyzed by NMR and UV-vis spectroscopy. The UV-vis spectrum of the product shows the formation of the technetium complex with acetylacetonate ligands. The NMR spectrum indicates that the product is diamagnetic and the acetylacetonate coordinates to technetium atom as a bidentate ligand. The product is probably the technetium(IV)-acetylacetonate complex having the oxo-bridged dinuclear structure.

1P01: Spectroscopic measurement of visible and ultraviolet light originating from de-excitation of ^{229m}Th

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An isomer of ^{229}Th (^{229m}Th) has on extremely low energy level. The de-excitation of ^{229m}Th is expected to occur via an electronic bridge mechanism in addition to a direct γ -ray transition. The decay process depends on its chemical condition because electronic bridge

mechanism is a process providing a part of de-excitation energy to excite the outer-shell electrons. We have performed experiments for the detection of visible and ultraviolet photons emitted from $^{229\text{m}}\text{Th}$ with a non-spectroscopic photon measurement system with low-background and high-detection efficiency. However, the spectroscopic measurement is essential to discuss the decay process of $^{229\text{m}}\text{Th}$. In this work, we have developed a photon spectrometer system for spectroscopic measurement for $^{229\text{m}}\text{Th}$ sample.

1P02 : Basic study of the mass spectrometry for gas phase chemistry

Ikezawa, T., Goto, S., Kudo, H., (Fac. of Sci., Niigata Univ.)

Rapid gas phase separations have been applied for the study of chemical properties of transactinide elements. In general, the chemical species involved were ambiguous in the gas phase separation. For the identification of chemical species, a mass spectrometry is considered by use of resonance enhanced multi-photon ionization (REMPI) method. In this work, the resonance wavelengths of dipivaloylmethane (DPM) currently used in the gas phase chemical experiment are searched with a tunable laser.

1P03 : Cation exchange behavior of Zr and Hf fluoride complexes— Model experiments for chemical characterization of superheavy element rutherfordium

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In order to determine the fluoride complex formation constants of rutherfordium, cation exchange behavior of the lighter homologues, Zr and Hf, in HClO_4/HF mixed solutions was studied as a model experiment. The distribution coefficients (K_d) of Zr and Hf in 3 and 4 M $\text{HClO}_4/0\text{--}10^{-2}$ M HF mixed solutions on the cation exchange resin were measured with a batch method using radiotracers ^{88}Zr and ^{175}Hf . It was found that the K_d values of Zr steeply decrease compare with those of Hf, indicating the stronger interaction between Zr and F. The K_d values of Zr and Hf entirely decreased with an increase of $[\text{HClO}_4]$. In the symposium, we also discuss the experimental results on the kinetics of the fluoride complex formation of Zr and Hf.

1P04 : Basic study on anion-exchange experiment for bromide complex of group-4 elements

Goto, S.,^a Hirose, H.,^b Sato, H.,^b Kudo, H.,^b (^aCenter for Instrumental Analysis, Niigata Univ., ^bFac. of Sci., Niigata Univ.)

So far, the anion-exchange behavior of the fluoride and chloride complex of the group-4 elements, Zr, Hf, and Rf, has been investigated. The orders of the K_d values were $\text{Rf} \ll \text{Hf} \approx \text{Zr}$ for the fluoride complex and $\text{Hf} < \text{Zr} < \text{Rf}$ for the chloride complex. For the bromide complex of Zr and Hf, the K_d values were measured using 1.0 – 9.0 M HBr. The K_d values were very small and were not so much different. Bromide ion concentration becomes up to about 9 M with HBr solution, but becomes up to 14 M with lithium bromide solution. The ion-exchange experiments (batch and column methods) were performed with LiBr solution using ^{88}Zr and ^{175}Hf as a tracer. As a preliminary result, the K_d values in the batch experiment increased with the increase in bromide ion concentration.

1P05 : Structural Study of Nb and Ta in HF Solution for Chemical characterization for Dubnium

Akiyama, K.,^a Tsukada, K.,^b Asai, M.,^b Haba, H.,^c Sueki, K.,^a Toyoshima, A.,^b Yaita, T.,^d Nagame, Y.,^b (^aGrad. School of Pure and Appl. Sci., Univ. Tsukuba, ^bAdv. Sci. Res. Center, JAERI, ^cCyclotron Center, RIKEN, ^dSyncro. Rad. Res. Center, JAERI)

We studied molecular structure of the metal complexes for Nb and Ta in HF solution using the EXAFS method to discuss the anion-exchange behavior of Db. The fluoride salt of Nb and Ta were dissolved in HF solution. Parts of the solution were mixed with the anion-resin for the investigation of chemical forms in the resin. These samples were set to the BL27B of PF at KEK. The radial structure functions (RSF) of Nb in solution were gradually changed with the increase of HF concentration but that in resin were dramatically changed between 10 M and 14 M. This trend in RSF is consistent with that in the adsorption behavior to anion-resin. The RSFs of Ta in solution is changed in the intensity over 1 M, but that in resin is not changed between 0.1 M and 26 M.

1P06 : Development of the gas-jet transport system for the superheavy element chemistry using the RIKEN AVF Cyclotron

Takabe, T.,^a Kitamoto, Y.,^a Saika, D.,^a Matsuo, K.,^a Tashiro, Y.,^a Yoshimura, T.,^a Haba, H.,^b Kaji, D.,^b Shinohara, A.,^a (^aGrad. School

Sci., Osaka Univ., ^bCyclotron Center, RIKEN)

We have developed a gas-jet coupled target system for production of the superheavy elements and a rotating wheel system for measurement of their alpha and spontaneous fission decays to start up the superheavy element chemistry using the RIKEN K70 AVF Cyclotron. The performance of the gas-jet system was investigated systematically using the W isotopes produced in the ²²Ne induced reaction on the ^{nat}Gd target as a function of the temperature of the KCl aerosol generator, the He-gas flow rate, and the ²²Ne⁷⁺ beam intensity. The background condition in the alpha measurement was evaluated with the rotating wheel system.

1P07 : Distribution coefficients measured with non-carrier tracers of Zr-88 and Hf-175 on TBP-Resin in hydrochloric acid and nitric acid solutions

Kamataki, S.,^a Morishita, K.,^b Haba, H.,^c Yokoyama, A.,^a (^aGrad. School Nat. Sci. Tech., Kanazawa Univ., ^bFac. Sci., Kanazawa Univ., ^cCyclotron Center, RIKEN)

Distribution coefficients of Zr and Hf were measured with non-carrier tracers of Zr-88 and Hf-175 on TBP-Resin in hydrochloric acid and nitric acid solutions in order to research for the chemical behaviors of heavy elements like Rutherfordium in solvent extraction system. The commercial resin was used as a support material for solvent of tributylphosphosphate. The tracers were produced in the bombardment of protons at Research Center for Nuclear Physics, Osaka University. The batch method was applied to the measurement by using the tracers. The results show that the measured values are larger than the data in a reference in the ordinary concentration by one order and they have similar tendency as functions of the acid concentrations of HCl and HNO₃. It was concluded that the solution system of nitric acid in 6 to 8 M may be promising for the measurement of Rf.

1P08 : Development of detector to measure atomic number of fission fragments in on-line measurement

Sakamaki, M., Goto, S., Kudo, H., (Fac. of Sci., Niigata Univ.)

In order to investigate the mechanism of asymmetric mass division in low energy fission of actinides, atomic number (*Z*) of fission fragments are decided to be measured together with masses and kinetic energies. *Z* will be decided from the information of energy losses of fragments. For this purpose the gridded ionization chamber with divided anode is designed and tested.

1P09 : Measurement of etch pit profile in CR-39 with an atomic force microscope

Arai, M.,¹ Yokoyama, A.,¹ Hirano, T.,² Matsumura, H.,³ (¹Graduate School of Natural Science and Technology, Kanazawa Univ., ²Graduate School of Science, Kanazawa Univ., ³High Energy Accelerator Research Organization)

In measuring nuclear reaction products using a solid-state nuclear track detector, improvement of nuclide identification from information on etch-pit will make the method more useful in many research fields. By using a CR-39 we investigated the relationship between nuclear reaction products and the information on the etch-pits observed with an atomic force microscope for the products of nuclear fragmentation induced by heavy ion. But the measurement of etch-pits was very difficult for damage of the detector caused by incident particles. Besides, we are devoid of standard data to identify such products. In this study, we performed irradiation using high-energy neutrons as incident particles and also an experiment for standard data by implanting 25 MeV ¹²C particles directly to CR-39. Although the background tracks were found, we obtained a result showing possibility that only the fragmentation product is recognized if the detector is etched for a short time. The standard data were subjected to reanalyzing the fragmentation data taken before.

1P10 : Evaluation of neutron self-absorption in gold and application to measurement of neutron flux

Murata, Y.,^a Komura, K.,^a Koga, T.,^b Morishima, H.,^b (^aLLRL, Kanazawa Univ., ^bAtomic Energy Res. Inst., Kinki Univ.)

In order to use gold as excellent neutron detector, self-absorption of neutrons in gold target was experimentally evaluated by using Kinki University nuclear reactor (UTR-KINKI). Piled up thirty gold plates, sizes of which were 10 × 10 × 0.15 mm, were irradiated for 10 minutes by neutrons in center of UTR-KINKI (neutron flux: 1 × 10⁷ n·cm⁻²·s⁻¹). Ten gold plates, sizes of which were 40 × 30 × 0.2 to 0.7 mm, were irradiated by leakage neutrons for 6 hours at outer wall of UTR-KINKI (neutron flux: 10 n·cm⁻²·s⁻¹). The ¹⁹⁸Au activities in the gold plates were measured by γ -ray spectrometry. Relative ¹⁹⁸Au activities decreased considerably within 1 mm from both sides of the gold plates, but were almost constant over 1 mm. We consider the gold plates near center were influenced by neutron irradiation from both sides in the same. These data of neutron absorption will be useful for measurement of natural neutron flux by using gold detector, because natural neutron actually irradiate

from both sides of gold plates.

1P11 : Extractive behavior of actinide elements and development of an automatic liquid scintillation counting system for the α -ray detection.

Tashiro, Y., Kitamoto, Y., Saika, D., Matsuo, K., Takabe, T., Sato, W., Takahashi, N., Yoshimura, T., Shinohara, A., (Grad. School of Science, Osaka Univ.)

In order to investigate the chemical properties of heavy actinide elements, we studied about the solvent extracting behavior of some lanthanides and Am and Cm in water-benzene phase by using of some kinds of β -diketones (2-Thenoyltrifluoroacetone (TTA), 1,1,1,5,5,5-Hexafluoroacetylacetylacetonone (HFA), 4,4,4-Trifluoro-1-phenyl-1,3-butanedion (TPB)) as the extracting agents. We will also report about the approach for improvement of α rays and β rays discrimination on liquid scintillation counting including the development of the flow counting system using the liquid scintillation counter.

1P12 : Separation of the actinide elements by capillary electrophoresis and derivation of the ion radius.

Kitamoto, Y.,^a Saika, D.,^a Matsuo, K.,^a Takabe, T.,^a Tashiro, Y.,^a Sato, W.,^a Yoshimura, T.,^a Haba, H.,^b Enomoto, S.,^b Mitsugashira, T.,^c Shinohara, A.,^a (^aGrad. School. Sci., Osaka Univ., ^bCyclotron Center, RIKEN, ^cOarai-branch, Inst. Materials Res., Tohoku Univ.)

In the chemistry of heavy actinide elements, chemical isolation of a desired nuclide is required to detect one atom at a time. It must be performed rapidly, because of the short lifetime of them. Capillary electrophoresis (CE) enables us to separate them rapidly. At the same time, we can get some knowledge of the ion radius. Generally in CE, an analyte is detected by on-capillary detection for good separation. But we tried to collect fractions of the eluent containing the nuclides, because it is difficult to detect the elements in this way. A simulation experiment was performed using radiotracers of several lanthanide elements, which are lighter homolog of actinide elements. In this report, we describe the result of the experiment, and discuss the relation of the mobility to the ion radius of each element.

1P13 : Mössbauer and XAFS spectroscopic studies of (Y,Zr)-Np-O systems (2)

Nakada, M.,^a Otake, H.,^a Yamashita, T.,^a Akabori, M.,^a Minato, K.,^a

Motohashi, H.,^b (^aDepartment of Materials Science, Japan Atomic Energy Research Institute, ^bTokyo Nuclear Service)

Cubic stabilized zirconia and zirconium pyrochlores are useful for nuclear and non-nuclear fields. Ytria stabilized zirconia is the popular and promising material because of its wide range of stability in the phase diagram. The single phase of Np-doped yttria stabilized zirconia, $(Y,Zr)_{0.95}Np_{0.05}O_{2-x}$ and $(Y,Zr)_{0.8}Np_{0.2}O_{2-x}$ were confirmed by powder X-ray diffraction techniques. Extended X-ray absorption fine structure (EXAFS) spectra were collected on the Y, Zr K edge and Np L_{III} edge at the Photon Factory in the Institute of Materials Structure Science of the High Energy Accelerator Research Organization. The bond length of the Zr-O, Np-O and Y-O for $(Y,Zr)_{0.95}Np_{0.05}O_{2-x}$ were 2.210, 2.248 and 2.354 angstrom by analysis for 1st shell of EXAFS spectra, respectively. These bond length values were shorter than those for $(Y,Zr)_{0.8}Np_{0.2}O_{2-x}$. These results were agree with lattice parameters by powder X-ray diffraction measurements.

1P14 : Crystal structure of the Uranyl(VI) complex coordinated by both ligand N and O atoms

Kawasaki, T.,¹ Kitazawa, T.,¹ (¹Fac. Sci., Toho Univ.)

Single crystal of $[UO_2(NO_3)_2(terpy)]$ have been synthesized by adding the acetonitrile solution of the uranyl(VI) nitrate to the chloroform solution of the terpy ligand at room temperature. The crystal structure has been determined by using X-ray CCD. The $[UO_2(NO_3)_2(terpy)]$ complex exhibits hexagonal-bipyramidal geometry about the uranyl(VI) ion. The compound involves one tridentate terpy molecule and two nitrate ions. The one of the nitrate ion is monodentate, and another is bidentate.

1P15 : Solvent extraction of trivalent lanthanide and actinide using dioctyldithiocarbamate.

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The extraction behavior of ^{152,154}Eu(III) and ²⁴¹Am(III) in HNO₃ solution using dioctyldithiocarbamate(DODTC) - nitrobenzene were investigated. It was found that the distribution ratios of Am(III) in the region of 5<pH<7 is extremely higher than that of Eu(III) when the aqueous phase contains 1 M sodium nitrate. The slope of log D vs -log[H⁺] on Eu(III) and Am(III) are +3 in the region of pH>5.8 when initial concentration of DODTC is 0.51 M, but the slope of

Am(III) is +5 in the region of $5.3 < -\log[H^+] < 5.8$. The variation in the slopes of $\log D$ vs $-\log[H^+]$ in Am(III) is caused by the variation in the concentration of DODTC in organic phase. The concentration of DODTC in organic phase is dependent on $-\log[H^+]$, because the decomposition of DODTC by nitric acid contributes.

1P16: Extraction equilibrium of Am(III) and Eu(III) with tris(2-methylpyridyl)amine

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Recently finding new selective ligand on the extraction of Am(III) and Eu(III) has been growing interest. Several studies using soft-donor ligand containing nitrogen and sulfur atoms were reported. A soft-donor ligand, tris(2-pyridylmethyl)amine (TPA) which has tripodal three pyridylmethyl groups, forms stable complex with lanthanides [1]. On the extraction of Am(III) and Eu(III) using decanoic acid or picric acid in 1,2-dichloroethane and nitrobenzene, we found that the selectivity of Am(III) over Eu(III) is improved by the addition of TPA. The separation factors (Am/Eu) are 20-50. In this study, we have elucidated the extraction equilibrium of Am(III) and Eu(III) with decanoic acid or picric acid in the presence of TPA in 1,2-dichloroethane. We will have a presentation in terms of the equilibrium analysis.

1P17 : Solvent extraction of f-block element ions with thiacalix[4]arenes and their oxides

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It is expected that thiacalix[4]arenes and their oxide (sulfinyl- and sulfonylcalix[4]arenes) have the ability of selective complexation for metal ions, based on HSAB principle. The extraction behaviors of $^{152,154}\text{Eu(III)}$ and $^{241}\text{Am(III)}$ in the region $\text{pH}=4\text{--}6.3$ were investigated using those extractants dissolved into chloroform. Both metal ions were extracted by those extractants in a following order; sulfinyl- > sulfonyl- > thiacalix[4]arene. When the aqueous phase contains 0.1M acetic acid buffer, the sulfinyl- and sulfonyl compounds showed efficiently extraction for both elements, but thia compounds showed poor extractability. On the other hand, when the aqueous phase dose not contain acetic acid buffer, thia compounds also showed efficiently extraction for both metal ions.

1P18 : The interaction between trivalent f-block element ions and iodide ion

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The interaction of $\text{Eu}^{3+}\text{-I}^-$ and $\text{Am}^{3+}\text{-I}^-$ has been studied by a back-extraction technique in a mixed solvent of methanol and water at 1.0 mol dm^{-3} ($\text{H, Na}(\text{I, ClO}_4)$). It was revealed for the interaction to be a solvent-shared ion-pair and a solvent-separated ion-pair from the interionic distance between metal ion and iodide ion. In the methanol mole fraction (X_S) < 0.31, the formation constants of $\text{Eu}^{3+}\text{-I}^-$ ($\beta_1(\text{Eu})$) are somewhat larger than $\beta_1(\text{Am})$, but the values of $\beta_1(\text{Eu})$ and $\beta_1(\text{Am})$ at $X_S = 0.31$ and 0.40 are approximately same. The values of $\beta_2(\text{Am})$ at $X_S = 0.31$ and 0.40 were measured, the other hand, the $\beta_2(\text{Eu})$ were not measured. So, Eu(III) and Am(III) species at $X_S = 0.31$ and 0.40 have different average charge. This difference may be applicable to the separation of Eu(III) and Am(III).

1P19 : Study of metallofullerenols by radiochemical method

Iwai, Y., Sueki, K., (Grad. School. of Pure Appl. Sci., Univ. of Tsukuba)

Water-soluble fullerene derivatives have been investigated to date mostly within the framework of biological and medical applications. For example, C_{60} derivatives have been applied as HIV inhibitors, radical scavenging capabilities and anti-cancer. We investigate to trace the synthesis process of metallofullerenols using radioactivity, and obtain the effective process of radio-metallofullerenols. Isolated $\text{Sm}@\text{C}_{82}$ including ^{153}Sm with C_{60} in toluene were vigorously vibrated with saturation KOH aq containing few drops of TBAH (10 % in water) as catalyst at room temperature. The reaction times were changed from 1 minute to 8 hours. The reaction mixture was filtrated by $0.2 \mu\text{m}$ filter. The residue was rinsed with methanol to ensure the removal of the catalyst and KOH. The rinsed residue was dissolved in water for 7 hours. The resultant brown solution was passed down a Sephadex G25 size-exclusion gel chromatography column using distilled water as the eluent. All products of reaction process were traced 103.2 keV γ -emission of ^{153}Sm by HPGe detector. We report a time change tracer's yield in each phase and the overall product yields of $\text{Sm}@\text{C}_{82}(\text{OH})_n$.

1P20: Study on Hf metallofullerenes using ^{181}Hf radioactive-tracer

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Recently, we reported new Hf metallofullerenes of $\text{Hf}_2\text{C}_2@C_{78}$, Hf_3C_{82} , Hf_2C_{82} , Hf_2C_{84} and $\text{Hf}_2\text{OC}_{80}$. However, their structures and chemical properties have not been clarified because of their small amount of production yields. Here, we report the HPLC elution behavior of Hf metallofullerenes on 5PBB, Buckyprep, Buckyprep-M and Buckyclutcher columns by use of ^{181}Hf radioactive-tracer and discuss the chemical properties from the HPLC elution behavior of Hf metallofullerenes on each stationary phase. From the obtained radiochromatogram, the retention time of $\text{Hf}_2\text{C}_2@C_{78}$ was almost the same as that of C_{90} on a Buckyprep separation. In a 5PBB separation, the retention time of $\text{Hf}_2\text{C}_2@C_{78}$ was smaller than that of C_{90} based in a Buckyprep injection. While, remarkable delay in the retention time of $\text{Hf}_2\text{C}_2@C_{78}$ was observed on a Buckyprep-M and Buckyclutcher separation. Comparing the capacity factor of $\text{Hf}_2\text{C}_2@C_{78}$ to C_{90} on a Buckyprep-M with that on a Buckyclutcher, latter was slightly large. In the session, we will discuss the chemical properties of Hf metallofullerenes from the differences in the capacity factors on each column.

1P21 : Concentration of ^{188}Re solutions from alumina-based $^{188}\text{W}/^{188}\text{Re}$ generator

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Rhenium-188 is a useful generator-produced radioisotope for radiotherapy because of their energetic beta particles and gamma rays suitable for imaging. Because the ^{188}W parent is available in a relatively low specific activity from the irradiation of enriched ^{186}W in a reactor, relatively large volumes of normal saline are required for elution of the ^{188}Re daughter from traditional alumina-based $^{188}\text{W}/^{188}\text{Re}$ generators. In this study, simple method for effective concentration of ^{188}Re solutions from an alumina-based $^{188}\text{W}/^{188}\text{Re}$ generator was investigated by a tandem IC-H cation- QMA SepPak anion column system. 0.3 M ammonium acetate was used for the elution of ^{188}Re from the $^{188}\text{W}/^{188}\text{Re}$ generator. Because the ammonium cations are exchanged for hydrogen ions through the first IC-H column, ^{188}Re was adsorbed to the QMA SepPak without interfering from acetate anion. Rhenium-188 was eluted from the

QMA SepPak column with normal saline after washing with water. About 90% of ^{188}Re was recovered in 1 ml of saline.

1P22 : Variations of ^7Be concentrations in surface air at Nagano City during the last five years

Muramatsu, H., Wada, M., Ishii, T., Abe, T., Horiuchi, Y., Yoshizawa, S., Nakagami, K., (Shinshu Univ.)

The concentrations of ^7Be in surface air at Nagano City have been measured during the period from August 2000 to July 2005. The average monthly concentrations of ^7Be in surface air were in the range of 2.5-14 mBq/m³ with pronounced two peaks in spring and autumn, which is almost in agreement with those observed at Osaka, Chiba, and Tsukuba in Japan. When comparing the present data at Nagano City with those at these cities, the most remarkable difference is that in winter the values at Nagano City are apparently lower, which may be conjectured that this difference is mainly due to snowfall, that is, at Nagano City there is a lot of snowfall during winter, on the other hand, at Osaka, Chiba, and Tsukuba very little. Together with anticorrelation between events of precipitation and concentrations of ^7Be , it is confirmed that wet precipitation is one of the main and important factors for the variations of ^7Be concentrations in air, especially as a short-period reduction.

1P23 : High resolution Simultaneous Measurements of Airborne Radionuclides –Influence of Meteorological Conditions –

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By the use of extremely low-background Ge detectors at Ogoya Underground Laboratory (270 mwe), it became possible to detect extremely low levels of environmental radionuclides. In this study, we tried to measure high resolution simultaneous measurements of airborne Pb-210, Po-210 etc. at three monitoring points, i.e.; LLRL in Nomi City, Hegura Island located 50 km from Noto Peninsula and Shishiku Plateau (640m a.s.l.) located about 8 km from LLRL. Various interesting results on the concentrations and variation patterns of airborne radionuclides were obtained, particularly, during drastic meteorological changes such as the passage of typhoon or snow fall.

1P24 : Tritium distribution in river and lakes and effect of nuclear tests

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Tritium is formed from several interactions of cosmic rays with gases of the upper atmosphere. Nuclear facilities and nuclear weapons tests also produce the tritium. Nuclear weapons tests at 1960s released copious tritium for environment, but present environmental tritium has decreased and is returning to the level before a nuclear tests. The purpose of this study is measuring the background level of the environmental tritium in river and lakes at 2005. Moreover, the next one is evaluating the influence of the nuclear weapons tests at 1960s in tritium concentration. We measured the tritium concentration in major rivers and lakes in Japan.

1P25 : Tritium concentrations in environmental samples collected at Kagoshima Pref. –2000~2002–

Sakae, N.,^a Shirasaka, K.,^a Ohtsu, M.,^a Imamura, H.,^a Izumo, N.,^a Kawamura, H.,^b Fukuda, M.,^b Tagomori, H.,^b Hirai, E.,^b and Matsuoka, N.,^b (^aKagoshima Pref. Inst. Environ. Res. Pub. Health, ^bKEEA)

We measured tritium concentrations of various samples (HTO in the atmosphere, sea water, river water and biological samples) collected near PWR nuclear power plant and from general environment of Kagoshima Pref. in the periods from April, 2000 through March, 2003. We confirmed recent tritium level and evaluate the dynamics in the environment.

1P26 : Method for measurement of ¹²⁹I in soil sample by using AMS –Target preparation–

Kawamura, H., Tennichi, Y., Matsuoka, N., (KEEA)

We investigated AgI target preparation method for measurement of ¹²⁹I in soil sample by using AMS. The method is composed of the following three procedures: (1) iodine collection into alkaline solution by soil combustion, (2) iodine purification by extraction, (3) iodine collection as AgI by addition of AgNO₃. We evaluated iodine background level and recovery in the procedures.

1P27 : Insolubilization of Tc-99 by marine bacteria *Halomonas* sp.

Fujimoto, K., Minakawa, M., Morita, T., (National Research Institute of Fisheries Science)

We separated marine bacteria (TC202 strain), which had the capability to insolubilize technetium-99 (Tc-99; physical half-life = 2.111 X 10⁵ years) from the aqueous phase in aerobic condition. TC202 strain was identified as *Halomonas* sp. by sequencing analysis of 16S rRNA gene. TC202 strain was incubated in the PPES-II medium supplemented with 10 Bq / mL of Tc-99 (TcO⁴⁻) at 15°C for 24 hours. After centrifugation at 15000 rpm for 5 minutes, the radioactivity of Tc-99 in each fraction was measured by liquid scintillation counter. The amount of Tc-99 fractionated to the precipitation was 53.5 ± 3.2%. Paper chromatography analysis indicated that Tc-99 in the insoluble fraction was reduced from Tc(VII) to Tc(V). Insolubilization of Tc-99 by TC202 strain was inhibited by heavy metal ions (Fe³⁺, Cu²⁺, Ni²⁺ and Co²⁺) or chelating reagent (EDTA and EGTA).

1P28 : ^{239, 240}Pu inventories in marine sediments in the vicinity of the Izu-Ogasawara Trench

Kihara, S., Nishizawa, A., Chonan, Y., Nakanishi, T., (Grad. School Nat. Sci. Tech., Kanazawa Univ.)

The artificial radionuclides ^{239, 240}Pu ($T_{1/2}$ = 2.41 × 10⁴ y, 6.56 × 10³ y) in the environment were released from atmospheric nuclear tests carried out between 1945 and 1980. The ocean, by virtue of its large surface area, has received a major share of the fallout radionuclides. It is not well known about the sinking process of Pu from the sea surface to the sediment. In the present study, we measured Pu depth profile in a sediment, and the Pu inventory in the sediment column was compared with that in a water column. ^{239, 240}Pu inventories were determined in the sediment column to be ~1.7 mBq cm⁻², and in the seawater column to be ~4.6 mBq cm⁻². This indicates that ~27 % of fallout Pu reached on the sea-surface has buried in the sediment by the year 1994.

1P29 : Depth profile of radionuclides in a ferromanganese crust

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The depth profiles of radionuclides in a ferromanganese crust were measured to determine growth rate and to identify the contaminated depth with fallout nuclides as a part of a project to search for a supernova remnant. The ferromanganese crust used for this work was taken on a sea-mount (water depth of 1551 m) between Wake Island and Johnston Island in 1994. The crust with surface area of ~420 cm² and thickness of ~5 cm was sliced 5 mm intervals, and Be, Th, Pa, U, and Pu were chemically separated by means of a precipitation, solvent extraction, and ion exchange. ²³⁰Th, ²³²Th, ²³¹Pa, ²³⁴U, ²³⁸U, and ^{239, 240}Pu alpha-activity were measured with a Si semiconductor detector, and ¹⁰Be contents were measured with an AMS system. A growth rate of 2.3 mm/10⁶ y was evaluated from the depth profile of ¹⁰Be, and ^{239,240}Pu was found in the surface (0~0.5 cm) layer of the crust.

1P30 : External radiation in Dolon village due to fallouts from the Semipalatinsk Nuclear Test Site

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Dolon village, located about 110 km ENE from the ground-zero point in the Semipalatinsk Nuclear Test Site, was contaminated by fallouts from the first USSR atomic bomb test in 1949. The current level of Cs-137 soil contamination in Dolon was described by a log-normal distribution with an average of 4.2 kBq/m². A 90 percentile of this distribution was adopted as a referential contamination in Dolon, which corresponded to 32 kBq/m² in 1949. A VB Excel program, FPCOMP.xls was developed to follow temporal change of FP composition after nuclear explosions. Air-dose of 0.016 ± 0.004 Gy per 1 kBq/m² of Cs-137 deposition was obtained for the time period from 3 hr (assumed arrival time of radioactive clouds to Dolon village) to 1 yr after the explosion. Finally, combining this value with the Cs-137 deposition in 1949, external radiation of 0.51 ± 0.13 Gy was evaluated as the referential air-dose in Dolon village.

1P31 : Radioactivity content of papers (IV)

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The radioactivity of a fallout nuclide ¹³⁷Cs in papers such as magazines and newspapers produced in 1999-2000 in Japan was determined by gamma-ray spectrometry to obtain information on radioactivity level of papers. The average ¹³⁷Cs contents of weekly general interest magazines, weekly comic magazines, and

newspapers were respectively 0.95, 0.06, and 0.11 Bq kg⁻¹, whereas the radionuclide was not detected in the other kinds of samples. The ¹³⁷Cs contents of weekly general interest magazines were comparable to those of woods from trees that grew in the 1960s. A phloroglucinol color test showed that the main printed pages of the weekly general interest magazines were made up of mechanical pulp. Possibly the mechanical pulp kept ¹³⁷Cs that had been contained in its material wood. It is likely that in paper production in Japan, most of ¹³⁷Cs contained in material wood is lost by migrating to water solution. The radioactivity of ¹³⁷Cs that moves from wood to water solution in paper production in Japan was estimated to be 15 GBq.

1P32 : Temporal variation of Carbon-14 concentration in tree-ring cellulose for the recent 50 years

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Concentration of organically-bound ¹⁴C in the tree-ring cellulose of a pine tree grown in Shika-machi, Ishikawa prefecture, Japan (37.1°N, 136.5°E), was measured for the ring-years from 1949 to 1999 and compared with those in several trees from East Asia region reported by other researchers. The difference of Δ¹⁴C values was found during the period of 1970-1981 between our data and those of other reports, in addition to the difference during the period of 1963-1967 caused by the so-called latitude dependence of the ¹⁴C variations in the northern troposphere. These results suggest that the ¹⁴C concentration in atmospheric CO₂ was considerably disturbed during the period of 1970-1981, especially in 1970, 1976, and 1978-1981, in the East Asia region. This phenomenon may be interpreted by the possibility of the several times of injections of ¹⁴C originated from a series of Chinese thermonuclear bomb tests.

1P33 : Study of determination of radium in environmental water by barium carbonate coprecipitation/liquid scintillation alpha spectrometry

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To separate radium is necessary for determination of radium by liquid scintillation alpha spectrometry. Generally, it is done by a barium sulphate coprecipitation. However, the procedure of this method is complex. We investigated a barium carbonate coprecipitation method for easily radium separation and

determined radium in Arima hot spring water. Ra-224 (Half time is 3.66 days) in thorium nitrate solution was used as spike samples for a method validation. Recovery and reproducibility of radium separation procedure were obtained by an activity of Ra-224. As a result of the spike experiment, recovery of Ra-224 was able to get about 87% and relativity standard deviation was able to get 5%. In Arima hot spring water, Ra-226 is 3.6 ± 0.3 mBq/L, Ra-224 is 1.5 ± 0.1 mBq/L.

1P34 : High resolution simultaneous measurements of airborne radionuclides at sub-regional sampling points by ultra low background gamma spectrometry

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By using 11 extremely low background Ge detectors at Ogoya Underground Laboratory, it became possible to investigate temporal variations of airborne ^{212}Pb ($T_{1/2} = 10.6$ h) along with ^{210}Pb and ^7Be at time intervals of 2-3 hours. We have measured above radionuclides at three monitoring points viz, 1) Low Level Radioactivity Laboratory (LLRL) Kanazawa University, 2) Shishiku Plateau (640 m MSL) located about 8 km from LLRL to investigate vertical difference of activity levels, and 3) Hegura Island (10 m MSL) located about 50 km from Noto Peninsula in the Sea of Japan to evaluate the influences of Asian continent or mainland of Japan on the variation to the activity levels. Whereas variations of long-lived nuclides, ^{210}Pb and ^7Be , showed simultaneity among 3 monitoring points, short-lived ^{212}Pb showed time lags, which might be attributed to the difference in altitude and distance of these sampling locations.

1P35 : Depth profiles of environmental neutron flux in solids

Hamajima, Y., Komura, K., (LLRL, Kanazawa Univ.)

Depth profiles of environmental neutron fluxes in iron and lead have been measured by the activation method of gold foils. The targets were 15 to 25 g gold foils (40 x 300 mm) of 0.19 g cm^{-2} (0.1 mm) in thickness. Seven targets were inserted at 0 (two targets), 22, 55, 110, 165, 275 mm depth of piled up iron absorber plates. Seven targets were also inserted at 0 (two targets), 5, 10, 15, 20, 30 mm depth of lead absorber plates and blocks. The photo peak was measured for 4 - 5 days using four well - and three planer - type of the ultra low background HPGe-detectors in the OUL at the same time. The neutron depth profiles in iron and lead are almost the

same, decreasing in the ranges from 0 to 26, and 17 g cm^{-2} , respectively, and having been almost constant at the investigated range of depths between 26 and 216 g cm^{-2} in iron and between 17 to 34 g cm^{-2} in lead. A maximum has not been found in shallow depth.

1P36: Obstruction of Sm to the determination of ultra-low-level ^{152}Eu separated from a sample exposed to atomic bomb

Inoue, Y., Nomura, T., Izumi, H., Hosotani, R., Yokoyama, A., Nakanishi, T., (Grad. School Nat. Sci. Tech., Kanazawa Univ.)

We have undertaken the determination of the specific radioactivity of ^{152}Eu (half-life: 13.542 y) in a sample exposed to the Nagasaki atomic-bomb at a place 1595 m distant from the explosion point. A chemical separation to prepare europium-enriched sample was performed for a 7.8 kg sample, and low-energy photon spectrometry was carried out. In the spectrometry, 39-40 keV Sm X-rays emitted after electron-capture decay of ^{152}Eu was measured because of the highest sensitivity. During careful counting, we found an excess ^{152}Eu activity compared to that estimated by DS02. A reason is obvious: the europium-enriched sample contains Sm, and Sm generates fluorescence X-rays. Hence, mock-up samples were prepared to assess the obstruction due to the fluorescence X-rays of Sm to the accurate determination of ^{152}Eu . It was confirmed that the intensity of fluorescence X-rays of Sm increases in direct proportion to the alpha-activity in the mock-up samples.

1P37 : Analytical method of trace level ^{99}Tc in environmental sample by using TEVA disc

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The first commercial nuclear fuel reprocessing plant in Japan locates in Rokkasho Village, Aomori Prefecture, and is now under the uranium test. Since the plant will release small amount of ^{99}Tc to the ocean, the concentration of ^{99}Tc in seawater nearby the plant will possibly increase in the future. Since it is important to get background levels of ^{99}Tc in environmental samples around the plant site, we developed an analytical method for trace-level ^{99}Tc in environmental samples. A known amount of $^{95\text{m}}\text{Tc}$ was spiked to the sample, then Tc in the sample was pre-concentrated by using a TEVA disc (Eichrom Industries, US). After Mo and Ru were

removed with an anion exchange resin and a TEVA resin column, ^{99}Tc in the sample was measured by an ICP-MS. We adopted the procedure for the analyses of international standard materials, IAEA-375(soil) and IAEA-381(seawater), and obtained good results. Seaweed samples from the northern Japan were analyzed for ^{99}Tc , and results will be presented.

1P38 : Trace analysis of uranium, thorium and lead in the environmental samples –examination of the scheme of anion-exchange separation–

Miyamoto, Y., Saito-Kokubu, Y., Magara, M., Sakurai, S., Usuda, S., (JAERI-Tokai)

We attempted to the separation of trace amounts of uranium, thorium and lead from the safeguards environmental samples with an anion-exchange column to analyze the elemental concentration and isotopic composition. The ability to separate each of these elements was assessed with elution curves. A mixture of three calibration standards for ICP-MS was used for as a synthetic sample for these examinations. The anion-exchange resin, Muromac 1x8 (Cl-form, 100-200 mesh), were packed into a column (5.5 mm in diameter and height of 42 mm). Every 30 drops of effluents were collected and the elemental concentrations in the effluents were measured with an ICP-MS to draw a elution curve. It was difficult to separate each element in the sample with the mineral acid medium only. In the use of the mixed medium of nitric acid and alcohol, the separation characteristics was bad, and more than 20 columns volume of the eluant was essential for the separation of the uranium from the sample. The mixed medium of hydrochloric acid and alcohol gave us superior separation characteristics to that of nitric acid and alcohol. Ten columns volume of the eluant was enough to separate the uranium from the sample.

1P39 : Development of a rapid analytical method for uranium in urine.

Kuwabara, J., Watanabe, Y., Noguchi, H., (JAERI)

A rapid urinary uranium analytical method for monitoring of internal exposure was developed. To reduce required time for the analysis, an improved flow injection extraction chromatography system was joined to inductively coupled plasma mass spectrometer (ICP-MS). This system has two separation columns and can carry out two chemical separation processes in parallel. Wet digestion process by UV irradiation was performed for 2 ml of urine samples

with 0.2 ml of 30 % hydrogen peroxide. The digested samples were measured by the ICP-MS joined with the flow injection extraction chromatography system. The detection limit of uranium was below 1 ng/L-urine under the condition of analytical-grade reagents and ultra pure water. Although analytical time for chemical separation and uranium measurement was 7 min for one sample, the improvement using the dual column system practically reduced the analytical time to half (=3.5min). This method is very useful for the monitoring of internal exposure with the rapidness and low running cost.

1P40: Multi-elemental analysis of environmental samples collected for radiological analysis

Ishikawa, Y., Omokawa, K., Sato, N., (Environ. Radioact. Res. Inst. Miyagi)

Multi-elemental analysis of environmental samples collected for radiological analysis were carried out in Miyagi Prefecture by using an atomic absorption-method, an ICP-method and an XRF(x-ray fluorescence analysis)-method. Because of recent low concentrations of fallout-radionuclides in the environment, it is difficult to investigate distribution and behaviors of such nuclides. In this study, many elements in marine organisms were analyzed and the concentration differences were compared among species, in order to investigate analogies or correlations with radionuclides. Concentrations of heavy metal elements, namely Lead, Zinc, Iron and Manganese, in livers of abalone and soft tissues of oyster showed higher values compared to other organisms. Considering the probabilities of emission of such as ^{65}Zn , ^{59}Fe and ^{54}Mn into the environment due to nuclear explosion tests or peaceful utilizations of nuclear energy, it is interesting to study concentrations of above-mentioned elements in marine organisms.

1P41 : Sedimentary records of U and Th isotopes in bottom sediments from Lake Baikal.

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Variations of U and Th isotopes in sediment core obtained in 1998 from Academician Ridge in Lake Baikal were investigated to establish their sedimentary behaviors and to look for a linkage to paleoenvironmental changes. The depth profiles of ^{238}U contents and $^{234}\text{U}/^{238}\text{U}$ activity ratios in bulk sediments samples had large variations of 62.5-402.9 mB/g and 1.00-1.92, respectively, and

demonstrate the presence of autogenous ^{238}U in the bulk sediments. The ^{238}U in bulk samples were discriminated into the autogenous and lithogenous ^{238}U fractions. The variation of this autogenous ^{238}U displayed clear correlation with variations of Bi-SiO₂ content. However from findings on sequential leaching, U was concluded to be unlikely to accumulate to any large extent in the Bi-SiO₂ fraction. Indeed, the ^{238}U contents in carbonate plus Fe-Mn oxyhydroxides fractions were confirmed to be mainly responsible for the variation of autogenous ^{238}U . These samples are analyzed for more details with SEM and TEM now. This study highlights the potential use of lithogenous and autogenous U (Th) signatures in sediments to trace the behavior of U (Th) and to reconstruct environmental changes in the area surrounding the lake, including hydrology of lake catchments.

1P42 : Observation of adsorption behavior of fission-multitracer in soils

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Adsorption behavior of various elements in soils has been studied by means of instrumental analyses to elucidate the mechanism of radionuclide migration in the environment. However, it is difficult to observe the behavior of very trace elements in such ways. In this study, adsorption behavior of fission products was observed by using a fission multitracer prepared at KUR. The multitracer contains fission products with no carrier materials. In our previous study, it was found that using the multitracer is effective. The diluted multitracer solution (20 mL) and soil sample (2.0 g) were mixed in two weeks and separated by filtration. K_d values were obtained by measuring gamma-rays from the solution and the multitracer solution which was not mixed with soil sample. It was found that rare earth elements show very high K_d values for all soil samples, however, other elements, e.g. alkaline metals, show various values depending on the soil samples. The mechanism of adsorption behavior will be elucidated by considering differences of K_d values.

2A01: Spin-crossover iron(II) compounds with long alkyl chains

Hayami, S.,^a Hiki, K.,^a Inoue, K.,^b Maeda, Y.,^a (^aKyushu Univ., ^bHiroshima Univ.)

Liquid crystals are fascinating functional materials, and are

important in the field of advance materials such as electrooptic devices. The construction of metal-containing liquid crystals (metallomesogens) has recently attracted a great deal of attention. Novel physical properties may be discovered from the compounds with long alkyl chains, which will be promising in the field of material science. Based on this strategy, we have attempted to produce the iron(II) compounds with long alkyl chains, [Fe(C15-abpt)₂](BF₄)₂ (1), [Fe(1C16-bzimpy)₂](BF₄)₂ (2), [Fe(2C16-bzimpy)₂](BF₄)₂ (3), [Fe(3C16-bzimpy)₂](BF₄)₂ (4) and have succeeded in observing liquid crystal and LIESST effect. Furthermore LB film consisting of the compounds was prepared, and characterized by using Mössbauer spectroscopy.

2A02: Mössbauer spectroscopic study on the spin-crossover phenomena of the assembled complexes

Nakashima, S.,¹ Morita, T.,² Yamada, K.,² Inoue, K.,² (¹N-BARD, Hiroshima Univ., ²Grad. School Sci., Hiroshima Univ.)

Assembled complexes, Fe(bpa)₂(NCX)₂ (bpa=1,2-bis(4-pyridyl)ethane; X=S, Se, BH₃) enclathrated biphenyl, 2-nitrobiphenyl, 1,4-dichlorobenzene, or diphenylmethane. The skeletons are 1D, 2D grid, or interpenetration structure, depending on the anion and organic molecule. ⁵⁷Fe Mössbauer spectroscopy and SQUID measurement revealed that the present assembled iron complexes showed spin-crossover phenomena by enclathrating organic molecule, while the guest-free assembled complexes showed temperature-independent high-spin state. The ratio of spin transition and the transition temperature depended on the assembled structure.

2A03: Mössbauer spectroscopic studies of alkylammonium ferricyanide

Nakajima, Y., Katada, M., (Grad. School Sci., Tokyo Metropolitan Univ.)

[(C_nH_{2n+1})_mNH_{4-m}]₃[Fe(CN)₆] (m=1,2) has been prepared and investigated by Mössbauer spectroscopy, calorimetric measurements(DSC and TG-DTA) and powder X-ray diffraction. The compounds with n≥10 (m=1,2) have interdigitated layer structures. The interlayer separation of the compounds (m=2) was longer than that of the compounds (m=1) slightly. This indicates that the compounds (m=2) are not interdigitated fully. The quadrupole splitting (m=2) was bigger than (m=1). This indicates that the form of [Fe(CN)₆]³⁻ ion is influenced by differences of number of alkyl groups.

2A04: Mössbauer spectra and magnetization of dinuclear metal complexes with the bridging π conjugate ligands

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The dinuclear complexes of $[\text{Ni}_2(\text{tpa})_2\text{L}](\text{BPh}_4)_2$ and $[\text{Fe}_2(\text{tpa})_2\text{L}](\text{BPh}_4)_2$ [L = CA(chloranilic acid, DHBQ(2,5-dihydroxy-1,4-benzoquinone), OX(Oxalic acid), and tpa=tris(2-pyridylmethyl) amine)] are synthesized and characterized with Magnetic susceptibility, Mössbauer spectroscopy, FT-IR, powder X-ray diffraction(XRD), and the elemental analysis. The Mössbauer spectra at 80K of $[\text{Fe}_2(\text{tpa})_2\text{L}](\text{BPh}_4)_2$ indicate only one iron site with the typical high spin state Fe^{2+} . Variable-temperature magnetic susceptibility measurements of $[\text{Ni}_2(\text{tpa})_2\text{L}](\text{BPh}_4)_2$ [L=CA(1), DHBQ(2), and OX(3)] reveal the occurrence of relatively strong antiferromagnetic coupling. The exchange parameter J values are smaller than that for the corresponding $[\text{Ni}_2(\text{tren})_2\text{L}](\text{BPh}_4)_2$ (tren=tris(2-aminoethyl)amine). The order of the effective magnetic moment μ_{eff} is (3) > (1) > (2), same as the tren complexes.

2A05: Chemical reactions of laser deposited iron films with substrate materials.

Namiki, K.,^a Miyazaki, J.,^a Nomura, K.,^b Yamada, Y.,^a (^aTokyo University of Science, ^bThe University of Tokyo)

Iron films were deposited on various kinds of substrate by laser-ablation and were characterized by Mössbauer spectrometry. Laser-evaporated iron atom has high translational energy, which induces chemical reaction with a substrate material. YAG-laser light was focused by a convex lens onto a block of enriched ^{57}Fe metal in a vacuum vessel (10^{-5}Pa). The desired temperature of a substrate was kept at temperatures of 10 to 573 K using a closed-cycle helium refrigerator and a resistive heater. Conversion electron Mössbauer spectra (CEMS) of the deposited iron films were measured at room temperature using $^{57}\text{Co}/\text{Cr}$. Mössbauer spectra of iron films deposited on Co substrate at 10 K and 297 K have broad peaks because of deposited Fe atoms occupying various sites of Co solid. Mössbauer spectrum of the sample deposited at 573 K has sharp peaks, which indicate the formation of Fe-Co alloy (Fe atoms have homogeneous sites of Co solid). It was demonstrated that the laser-deposited Fe films form alloy with substrate materials, and that the reactions are dependent on the substrate temperatures.

2A06 : Substitution Effect and Magnetic properties of $\text{Sr}(\text{Ru},\text{Fe})\text{O}_3$ Perovskite

Nomura, K.,¹ Felner, I.,² Zboril, R.,³ Mashlan, M.,³ Kousaka, W.,¹ Oogoshi, S.,¹ Hashimoto, K.,¹ (¹The University of Tokyo, ²The Hebrew University, ³Palacky University)

CaRuO_3 (low spin Ru^{4+} , $4d^4:t_{2g}^4e_g^0$, $S=1$) shows the sharp change in ordering or spin glass behavior under 87K, and $\text{SrRu}_{1-x}\text{Fe}_x\text{O}_3$ doped with several % ^{57}Fe shows the behavior of metallic conductivity and ferromagnetism. The substituted $\text{Ca}_{1-x}\text{Sr}_x\text{RuO}_3$ has showed different magnetic properties. The magnetic properties of 2% ^{57}Fe doped $\text{Ca}_{1-x}\text{Sr}_x\text{RuO}_3$ prepared by a sol-gel method were compared with the bulk particles by XRD and Mössbauer spectroscopy. Magnetic properties and microstructures of $\text{Sr}(\text{Ru}_{0.5}\text{Fe}_{0.5})\text{O}_3$ doped with 5% Ca or Ba were further investigated by a SQUID, and Mössbauer spectroscopy. Mössbauer spectra of $\text{SrRu}_{0.5}\text{Fe}_{0.5}\text{O}_3$ were a little different from those of 2% ^{57}Fe doped in $\text{Ca}_{1-x}\text{Sr}_x\text{RuO}_3$. The iron state is closed to high spin Fe^{3+} ($S=5/2$). Ru ion is considered to be Ru^{5+} ($4d^3:t_{2g}^3e_g^0$, $S=3/2$), but the value of χT was smaller than the calculated values. The inner magnetic fields of Ca doped $\text{SrRu}_{0.5}\text{Fe}_{0.5}\text{O}_3$ show a little stronger than that of $\text{SrRu}_{0.5}\text{Fe}_{0.5}\text{O}_3$ and $\text{Ba}_{0.05}\text{Sr}_{0.95}\text{Ru}_{0.5}\text{Fe}_{0.5}\text{O}_3$ at low temperatures. A chemical pressure effect was observed in Mossbauer spectra.

2A07 : ^{197}Au Mössbauer spectra of cyclometallated digold(I) complexes with dithiolates

Kang, Y., Takahashi, M., Takeda M., (Dep. Chem., Toho Univ.)

Crystal structures of cyclometallated 1,1'-bis(diphenylphosphinoferrocene)digold(I) complexes with dithiolates are determined. In 1,3-propanedithiolato complexes, the $\text{Au}^{\text{I}}-\text{Au}^{\text{I}}$ distance is rather short as 302.8 pm due to aurophilicity while the distance becomes longer in the longer dithiolates; 707.0 pm in 1,6-hexandithiolate for example. The ^{197}Au Mössbauer spectra measured at 20 K shows that both isomer shift and quadrupole splitting values increase with increase in the number of bridging methylene groups. A good correlation between the Sham's σ parameters and the $^{31}\text{P}\{^1\text{H}\}$ NMR chemical shifts is observed.

2A08 : In-beam Mössbauer Spectroscopic Study of iron Species Produced in Neutron-irradiated Iron Disulfides.

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Matsue, H.,^g (^aRIKEN, ^bInternational Christ.Univ., ^cTokyo Univ. of Sci., ^dDaido Inst. of Tech., ^eTokyo Met. Univ., ^fOsaka Univ., ^gJAERI-Tokai)

Chemical and physical changes induced by nuclear reactions have been one of the research subjects of radiochemistry for several decades. Emission Mössbauer spectroscopy is a useful unique tool in investigating chemical and physical states of trace amounts of species formed by nuclear transformations in solids non-destructively. We have started a neutron in-beam emission Mössbauer spectroscopic study of ⁵⁷Fe formed via the ⁵⁶Fe(n, γ)⁵⁷Fe reaction in solid materials using a parallel plate avalanche counter (PPAC). In this paper we report some results of the study on neutron induced changes of iron species in a binary semiconductor (FeS₂). Two polymorphs of iron disulfide were investigated, one was pyrite having the rock salt type crystal structure, the other was marcasite with nickel arsenide structure. The in-beam emission Mössbauer spectrum of pyrite showed two Lorentzian doublets. One component corresponding to pyrite, and the other major component was tentatively assumed as Fe³⁺ at the interstitial site of pyrite. In case of marcasite, the spectrum was also composed of two doublets. In both iron disulfides, it was revealed that recoil of the cascade γ -ray emission promptly after the neutron capture reaction produced one new species distinguishable from the original target compound by in-beam ⁵⁷Fe Mössbauer spectroscopy.

2B01: Neutron activation analysis using k_0 standardization method

Oura, Y., Ebihara, M., (Tokyo Metropolitan Univ.)

Though instrumental neutron activation analysis using k_0 standardization method became popular in Europe, this method is not popular yet in Japan. We have been using the k_0 -method mainly for analysis of suspended particulate matters. For determination by k_0 -method, it is usual to utilize a commercial or homemade exclusive software. Recently IAEA developed a software for k_0 -INAA and opened without charge. We attempted to use k_0 -IAEA in order to compare with a commercial software (k_0 -DSM). After calibration of a Ge detector using calibrated ¹⁵²Eu and ¹³⁷Cs, non-calibrated ²⁴Na and some sources with single gamma-ray, some SRM samples were irradiated at JRR-4 for 20 min with some monitors (Zr, Lu, Au, and Fe). The determinations by k_0 -IAEA and k_0 -DSM were compared with certified values. Though determination values obtained by k_0 -DSM were consistent with certified values with $\pm 10\%$, those by k_0 -IAEA when samples were measured at close position were higher. And we found that

determinations by k_0 -IAEA were tend to be higher systematically as close in the detector. The possible reason is that calculated efficiency of the detector is incorrect, but is not clear yet.

2B02: Neutron activation analysis of cultivated oysters

Fukushima, M.,¹ Nakano, Y.,² Chatt, A.,³ (¹Ishinomaki Senshu Univeristy, ²Kyoto Univeristy Reactor, ³Dalhousie University)

Difference in trace element concentrations in oyster tissues cultivated in Ishinomaki (Miyagi, Japan) on hanging ropes at different sea depths were investigated using neutron activation analysis. Three groups of oyster were collected from a single rope at 1 m, 6 m, and 11 m depth. Two groups of organ, hepatopancreas and muscle, and gill and mantle were separated from soft tissues, washed in distilled water, freeze-dried, and pulverized. One portion of the powdered samples was irradiated for a short time at the Dalhousie University SLOWPOKE-2 reactor in Canada for Ag, Br, Cu, Mg, Mn, Na, Sc, and V analysis. Another portion was irradiated in Kyoto University Reactor in Osaka, Japan for Co, Cr, Fe, Rb, Sb, Sc, and Zn analysis. The organs of Korean oyster were also analyzed for comparison. Four kinds of organs, hepatopancreas, muscle, gill, and mantle were separated from soft tissues and analyzed same way as shown above. As a result, levels of most elements were found to be depth dependent. Details will be presented.

2B03: Effects on helium ion implantation for hydrogen isotope retention behavior in SiC

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Silicon carbide (SiC) was thought to be one of the future fusion contractual materials. In fusion reactor environment, hydrogen isotopes, helium and neutron with high energy will be implanted into SiC and it is important to evaluate hydrogen isotope, including tritium, retention and desorption behaviors from viewpoint of fusion safety. In this study, deuterium ions were implanted into SiC up to saturation and thereafter helium ions were additionally implanted. Effects on helium ion implantation for hydrogen isotope retention and desorption behaviors were studied by means of X-ray photo electron spectroscopy and thermal desorption spectroscopy.

2B04: Studies on hot atom chemical behavior of energetic ions in solids (VIII) ~Clarification of chemical behavior of energetic deuterium implanted into oxygen contained boron thin film~

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For D-T fusion devices, boronization is to be considered one of the candidate methods for the removal of impurities such as oxygen and carbon desorbed from first wall and wafted in vacuum vessel. Oxygen trapped in the boron thin film would be supposed to make a strong influence on the tritium retention and the trapping mechanism. In this study, oxygen contaminated boron thin films in various oxygen concentrations were prepared by the P-CVD and energetic deuterium ions were implanted into these films. The chemical states of films were evaluated by XPS and the deuterium desorption behavior was studied by TDS. From the B1s XPS spectra, two peaks which correspond to B-B bond and B₂O₃ bond were observed for oxygen contained boron thin film. After deuterium ions implantation, the chemical shift was found for B1s XPS spectra. For the TDS results, although two desorption peaks were found for pure boron thin film, three desorption peaks were observed for oxygen contained boron thin film. As oxygen flow rate was increased, deuterium retention was reduced.

2B05: Studies on hot atom chemical behavior of energetic ions in solids(VII) ~Chemical behavior of energetic deuterium implanted into tungsten carbide

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In the divertor region of fusion reactors, tungsten carbide (WC) layer might be formed by sputtering of tungsten and carbon, which will be used for the divertor and/or first wall materials, during long-term plasma discharge at high temperature. It is important to elucidate the tritium behavior in this layer from viewpoint of tritium safety of fusion reactors. In this study, chemical behavior of energetic deuterium implanted into WC was studied by means of X-ray photoelectron spectroscopy (XPS) and thermal desorption spectroscopy (TDS). The TDS spectra showed that the implanted deuterium was desorbed in the temperature region of 300-700 and 900-1100 K. From the comparison with our previous experimental results for HOPG, each was suggested to be attributed to desorption

of deuterium trapped by tungsten and carbon in WC, respectively. Although the amount of deuterium trapped by tungsten increased as the deuterium ion fluence increased, that trapped by carbon remained almost unchanged. These deuterium trapping mechanisms were discussed with taking account of XPS and TDS results.

2B06 : Correlation Between Thermal Annealing Behavior of Radiation Defects and Tritium Release Behavior in Neutron-Irradiated lithium meta-silicate

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In tritium blanket systems in D-T fusion reactors, energetic tritium was produced by the reaction of ⁶Li(n,α)T. To establish tritium recovery systems and tritium safety in fusion reactors, it is important to reveal hot-atom chemical behavior of tritium bred in the tritium breeding materials. In the present study, Li₂SiO₃ was used as the sample, which is one of the candidates for the solid tritium breeding materials. The annihilation processes of defects generated in neutron -irradiated Li₂SiO₃ were expected to have correlation with the chemical behavior of tritium produced in the sample. The irradiation defects and their annihilation processes were observed by means of ESR (Electron Spin Resonance). The existence of E'-center, which is an oxygen vacancy occupied by one electron, was suggested and the temperature range of its annihilation was found to be approximately 400-600 K. In the presentation, we will discuss the details of kinetics of the annihilation processes of the irradiation defects.

2B07 : Ortho-positronium annihilation processes in supercritical fluids

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We measured positron annihilation time spectra in N₂O and CO₂ in the wide range of density including gas, supercritical fluid and liquid phase. The time spectra were resolved into four components of positron lifetime in the medium. The first and the second longest lifetimes were ortho-positronium component. We conclude that ortho-positroniums are in the two different environments. The longest component was well described with the positronium bubble model and the ortho-positronium existed far away from positron super where some chemical species were formed by the positron irradiation. On the other hand, the lifetime of the ortho-positronium

near the positron super was shortened by the chemical species. We measured annihilation time dependence of Doppler broadenings of the annihilation gamma ray. In the case of N_2O , the broadening of the second longest component was similar to that of the para-positronium component. The spin conversion reaction with NO may take place in the N_2O media. In the case of CO_2 , the broadening showed a feature of the ortho-positronium pick-off annihilation. The chemical reaction with O^- may take place in the CO_2 media.

2B08 : Perturbed angular correlation studies of ZnO

Itsuki, Y., Sato, W., Takahashi, N., Shinohara, A., (Grad. School Sci., Osaka Univ.)

Zinc oxide has many interesting physical properties, which is known to originate from an excess of the metal component. However, there is still controversy whether the metal excess is due to the presence of oxygen vacancies or interstitial zinc atoms. To obtain microscopic information about the relation between the physical properties of ZnO and the creation of the deficiency by impurities, we have observed hyperfine field at 111-Cd probes by the time differential perturbed angular correlation technique. Different electric field gradients were observed for In-doped ZnO compared with undoped ZnO. In the case of In-doped ZnO, temperature dependence of the electric field gradient was observed. This indicates the change of the charge distribution surrounding the probe nuclei by doping Indium impurities.

3A01 : Influence of pre-OSL measurement on thermoluminescence glow curves

Yonezawa, Y.,^a Hashimoto, T.,^b (^aGraduate School of Science and Technology, Niigata University, ^bFaculty of Science, Niigata University)

Changes of thermoluminescence (TL) properties in natural, synthetic and their thermally annealed quartz samples were investigated with regard to bleaching effect due to pre-optically stimulated luminescence (OSL) measurement. As a result, glow curves associated with 200-335 °C violet TL (VTL) peaks significantly decreased by blue-light exposure of OSL for 200 s, while red TL (RTL) peaks showed no effect for the OSL-blue light illumination. Therefore, optically stimulated luminescence (OSL) sources found to be intimately related to VTL peaks rather than BTL ones. On the basis of these results, simultaneous TL- and OSL-dosimetries were examined using same quartz aliquot.

Evaluated RTL-doses from pre-OSL measurement gave good agreement with that from as-received RTL ones, while the doses from VTL were underestimated. Consequently, simultaneous RTL- and OSL-dosimetry techniques using same aliquot were confirmed to be useful tools as well as a cross check method when a little quartz sample was extracted from archaeological burnt specimens.

3A02 : Dependence of optically stimulated luminescence (OSL) signals on dose-rates in natural quartz

Takeuchi, T.,^a Hashimoto, T.,^b (^aGraduate School of Science and Technology, Niigata University, ^bFaculty of Science, Niigata University)

Dose-rate effects on the optically stimulated luminescence (OSL) signals have been studied for volcanic and hydrothermal origin quartz samples. Each grain sample was irradiated with X-ray of 30 Gy at different dose-rates. In high dose-rate (3.0 ~ 9.0 Gy / min), OSL signal increased with increasing dose-rates, while no dose-rate effects were observed in low dose-rate (~ 3.0 Gy / min) ranges. Contrary to the OSL signal, integrated radioluminescence (RadL) intensities, having emission peak of 400 nm, decreased with increasing dose-rates. As a result, OSL signals showed inverse proportion to RadL intensities. Thus, the dependence of dose-rates on OSL signals, in high dose-rate, could be attributed to the self-bleaching effects due to RadL signals, since the trapped electron could be bleached by not only the RadL from quartz grain themselves, but also by RadL from the ambient quartz grains, during irradiation. Consequently, it was confirmed that the quartz samples should be irradiated with dose-rate as low as possible in the OSL dating protocol.

3A03 : Luminescence dating for burnt stones from archaeological remains

Iba, T.,^a Tamaki, M.,^b Hashimoto, T.,^b (^aGraduate School of Science and Technology, Niigata University, ^bFaculty of Science, Niigata University)

Thermoluminescence (TL) and optically stimulated luminescence (OSL) from quartz or feldspar grain samples have been applied to dating of burnt archaeological materials. Practically, some burnt stones were collected from ancient fireplace sites, which might be used 10,000-35,000 years ago. Among burnt materials, the burnt stones are preferable for the TL dating because they are not generally so precious materials. Red (RTL)- and

OSL-measurements of quartz extracts and far-red TL (far-RTL) of feldspar extracts were used for the evaluation of naturally accumulated doses. Single-aliquot regenerative-dose (SAR) protocol was employed for each luminescence dating. As a result, naturally accumulated doses from RTL-measurements showed the highest values among luminescence measurements, such as OSL and far-RTL. This showed reflect nature of the unstable luminescence source and/or anomalous fading effects for OSL and far-RTL sources. On the basis of these results, the evaluated RTL dates were in good agreement with the estimated ages. In some case, the evaluated RTL were overestimated due to incomplete zero-setting of RTL signals. It was concluded that the RTL-dating results from quartz extracts are the most suitable for the archaeological burnt stones.

3B01 : Evaluation of coprecipitation with trace element on carbonate mineral by solid solution model

Yoshida, Y.,^a Yoshikawa, H.,^b Sato, T.,^c (^a NESI, ^b JNC, ^c IDC)

In the geological disposal system radium released from vitrified high level radioactive waste would be incorporated into carbonate minerals through the coprecipitation reaction even if the concentration of radium is less than solubility of a pure solid (e.g. $\text{RaCO}_3(\text{S})$). One of the dominant carbonate minerals is calcite in the deep underground, and the behavior for this mineral to take radium into its body was observed by experiment. Therefore, the coprecipitation reaction of radium with calcite was examined to evaluate radium solubility. For the evaluation of the property of Ra intake with calcite solid solution reaction is defined as a dominant mechanism because calcite does not have effective sites for ion exchange and surface complexation and in addition uptake of trace element is mainly observed at growth face which implies that coprecipitation is achieved by the atomic arrangement at growth face. Therefore experiment to precipitate calcite with trace element was performed and solid solution equation was derived. From the Gibbs free energy obtained from experiment, the excess free energy which means the deviation from ideal solid solution decreases with the trace element amount in the solid decreasing. This tendency is expressed by Margules model.

3B02 : Long-term corrosion data acquisition by Xray-CT of archeological iron for radioactive waste disposal study

Yoshikawa, H., Honda, T., (JNC, Tokai, Hitachi Engineering Co., Ltd.)

For long-term stability evaluation of artificial barrier materials to use for disposal of high-level radioactive waste, we carried out the natural analog study that assumed a similar process in natural environment. By a laboratory experiment, there is disagreement between researcher for the effect of corrosion product on long corrosion behavior for about 1,000 years. In this report, we introduce some long-term corrosion data for archeological iron, which has been buried for the about 1,000 year, as a natural analog study. We carried out non-destruction analysis with X-ray computed tomography, for the analysis of iron corrosion thickness of the rust layer. For a sample deposited for a long term in a weakly oxidising or reducing environment, the corrosion (depth) was 0.2 mm at the maximum in the 1,500 year. We obtained more same samples and found that the corrosion depths were less than 2mm in about 1,000 year.

3B03 : Brain regional uptake of manganese in the development of 6-hydroxydopamine-induced parkinsonian rats

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The regional accumulation rates of ^{46}Sc , ^{54}Mn , ^{65}Zn and ^{86}Rb were determined in 15 sections at the level of the substantia nigra of the brain 6-OHDA-treated parkinsonian brains and compared with those of control brains one and two weeks after 6-OHDA treatment, using a radioactive multitracer. As a result, one week after treatment the high Mn accumulation was markedly observed in the 6-OHDA-lesioned substantia nigra, striatum, globus pallidus and hypothalamus regions in comparison with the other elements. 6-OHDA causes auto-oxidation and rapid accumulation of Mn at the same time in this stage. Manganese has four difference roles in the development of Parkinson's disease. Manganese accumulation may induce other elements accumulations and stimulate dopamine auto-oxidation in early stage, and may scavenge free radicals as Mn-SOD two weeks after treatment. Manganese may be trigger of 6-OHDA lesioning to induce terminal injury and neuronal cell loss.

3P01: Relationships between radiation-induced luminescence properties and impurities on synthetic quartz

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Blue-thermoluminescence (BTL) in quartz was emitted by recombination between electrons from trapped center and Al-hole centers. Aluminum, which substitutes for silicon, is a major impurity in quartz. A negative relationship between the Al-impurity contents and BTL intensity is often observed in different portions of hydrothermal-origin quartz. In a previous study, it was confirmed that hydrogen radicals could operate as killers of Al-hole centers. However, the detail correlations and interactions among Al-impurity, BTL intensity and hydrogen radical are still unknown. These correlations are investigated using synthetic quartz, which contains adjusted impurity contents. First of all, slice samples were photographed TL color image (TLCI) after γ -ray irradiation. Subsequently, four parts (+X, -X, S, Z part) were compared in terms of TL, IR spectrometry and ESR measurement. TL intensities were negative correlation for Al-OH absorption. At low temperature irradiation, BTL was appeared at -180°C . Al-hole center signal and hydrogen center signal from ESR decreased at same temperature region. Hydrogen radical were recombined to Al-hole centers accompanied with BTL emission at -180°C .

3P02 : Sensitivity changes of radiation-induced luminescence dependent on Al impurity contents

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The radiation-induced luminescence properties, including differences of luminescence sensitivity and thermoluminescence (TL) coloration (red-TL; RTL and blue-TL; BTL), were investigated with respect to seven kinds of impurity contents (Al, Ti, Li, Fe, Mg, Cu and Ge) and the α - β phase inversion break temperatures. Some natural and synthetic quartz specimens over 30 kinds were collected from large variety of sources. Concerning the inversion temperatures, all natural samples exhibited lower inversion temperatures, ranging from 570.8 to 572.8°C , than that from pure synthetic quartz (573.0°C), since high Al impurity contents in quartz bring on low inversion temperatures. Particularly, the inversion temperatures of RTL-quartz moved toward much lower temperature side in comparison with BTL-ones. Consequently, two RTL- and BTL-quartz specimens were found to be classified at a boundary temperature of 572.0°C . The BTL sensitivities from natural and synthetic quartz samples showed a negative relationship with Al contents beyond about 20 ppm. Contrary to the BTL result, RTL sensitivity steeply increases as a curve of second degree-like function of the Al contents. It is

strongly suggested that two Al impurities within a short distance co-operate as RTL hole center.

3P03 : Milling effects on thermoluminescence of quartz

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We often have to crush/mill materials in question to extract quartz grains for thermoluminescence (TL) dating. Their surface layers (usually several tens μm), where might be affected by α -rays and crushing/milling, are etched out just in case. Therefore, we actually do not have enough knowledge how α -rays and crushing/milling cause effects on TL. To inspect milling effects on TL of quartz quantitatively, TL spectrum measurements were conducted using various sizes of milled grains (5 – $280 \mu\text{m}$) that were irradiated with 1 kGy of γ -ray from ^{137}Cs before the milling procedure. All sample had some TL peaks, and their peak intensities decreased with decrease of the grain diameter (20 – $280 \mu\text{m}$). Especially, they sharply decreased in finer grain diameter ranges ($< 20 \mu\text{m}$). This sharp decrease is due to surface TL resetting, of which the thickness of the surface layers is estimated to be 500 nm. From present study, we can conclude that milling effects on extract quartz grains are almost negligible whenever coarse grains ($\sim 100 \mu\text{m}$) are employed for TL dating.

3P04 : Changes of radioluminescence properties from quartz during radiation-irradiation

Shimizu, N., ^a Hashimoto, T., ^{a,b} (^a Grad. School Sci. Tech. Niigata Univ., ^bDep. Chem., Fac. Sci. Niigata Univ.)

Light emission phenomenon, so called radioluminescence (RadL), is observed from quartz during X-ray irradiation. Some RadL properties of natural and synthetic quartz at room temperature were compared as well as between different thermoluminescence (TL) quartz samples. Typical quartz samples exhibiting blue-TL (BTL-quartz) showed blue-RadL (B-RadL) and violet-RadL (V-RadL), which rapidly decrease down to low intensity levels during X-ray irradiation. On the contrary, red-RadL (R-RadL) from red-TL (RTL) quartz gradually increased and reached to saturation for longer irradiation while V-RadL behavior was in slight concordance with the RadL of BTL-quartz. In particular, B-RadL emission tendency of BTL-quartz was investigated from two viewpoints; the theoretical relationship between decrease of B-RadL

intensity and BTL dose-response curve, and the changes of B-RadL behavior measured at various temperatures. Because of B-RadL decrease gave a good correlation with BTL dose-response curve in theoretical treatment, B-RadL emission should be derived from the same site of BTL. That is, the decreasing tendency of B-RadL should be dependent on the decrease of B-RadL centers based on formation of BTL sources during radiation-irradiation.

3P05 : RTL (red thermoluminescence)-dating using quartz grains extracted from archaeological burnt materials in Yayoi-period

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When radiation-exposed quartz is heated, so-called thermoluminescence (TL) is detectable. Since the TL-intensity is proportional to accumulated doses of the quartz, dating of burnt archaeological materials could have been realized. In previous papers, our laboratory has reported that red-TL (RTL)-dating results using quartz grains extracted from Jomon pottery pieces were in good agreement with their manufacturing dates. In this study, we have applied RTL-dating to the younger samples (Yayoi-remnant place), including jar pieces used for burial (Kamekan) and pottery ones from the Yoshinogari site. Quartz grains were extracted from inner portions of the samples, followed by the estimation of naturally accumulated doses by RTL measurements in combined with the single-aliquot regenerative-dose (SAR) method. Annual doses were estimated from sample pieces as well as the surrounding soil by γ -ray spectrometry. RTL-dating results of some jar pieces used for burial were concordant with those from ¹⁴C-dating and predicted ages.

3P06 : Speciation of uranium series nuclides in core samples at Kanamaru, Niigata Prefecture

Kanai, Y., Kamioka, H., Watanabe, Y., (Geological Survey of Japan, AIST)

In order to elucidate the movement-retention behavior of uranium in the sediment, the uranium series nuclides such as U-238, U-234 and Th-230 in the boring core at Kanamaru, Niigata Prefecture, was studied using selective chemical leaching techniques. Two boring cores of 37.5m and 30.0m length were obtained. Uranium was rich in the core of 10-12 m depth from the surface and three selected samples were subjected to the successive leaching. The procedure contains (1) 1M AcONa / AcOH, (2)

Ammonium oxalate / oxalic acid (TAO reagent), (3) Na citrate / NaHCO₃ / Na dithionite, (4) H₂O₂, (5) HCl, (6) HNO₃ / HClO₄ / HF treatments. The uranium series nuclides in each leached fractions were determined by alpha spectrometry. The results indicate that U in AcONa/AcOH soluble and H₂O₂ soluble fractions are accumulated. On the other hand, U in TAO(oxalate) soluble and CBD soluble fractions are depleted. Uranium might be adsorbed to the sediment weakly and it is suggested that iron compounds in the sediment play an important role in this area.

3P07 : ²³⁰Th/²³⁴U activity ratio in the 1707 products from Fuji volcano

Kurihara, Y., Takahashi, M., Sato, J., (Meiji Univ.)

Observation was carried out on the radioactive disequilibrium between ²³⁴U and ²³⁰Th in the 1707 products from Fuji volcano which is locating along Izu-Mariana island-arc, in order to discuss on the radioactive disequilibrium between ²³⁴U and ²³⁰Th and their bearing on problems such as mechanism of magma genesis, transfer of magmas towards the surface and magma dynamics. Thorium and uranium in the rock samples were separated by anion-exchange resin and purified by TEVA-Spec. and UTEVA-Spec. resins, respectively. Purified Thorium and uranium were electrodeposited onto a stainless steel planchet for alpha-ray counting. U-234 and ²³⁰Th in rock samples were determined by isotope dilution method coupled with alpha-ray spectrometry. Activity ratio of ²³⁰Th/²³⁴U in the 1707 products from Fuji volcano ranged from 0.93 to 0.71. The 1707 products from Fuji volcano were found that ²³⁴U is enriched relative to ²³⁰Th, ²³⁰Th/²³⁴U activity ratio < 1, which is often observed for volcanic products from subduction zones.

3P08 : ²³⁰Th/²³⁴U activity ratio in volcanic products from Kozu-shima and Niijima

Takahashi, M., Kuribara, Y., Sato, J., (Meiji Univ.)

Radioactive disequilibria among U-series nuclides in erupting magmas have been investigated with products from volcanoes in localities along the subducting zone, on the hot-spot or along the mid-oceanic ridge. With 45 volcanic rocks erupted recently from 6 volcanoes located along Izu-Mariana island-arc, Japan, ²³⁴U is reported to be in radioactive equilibrium with ²³⁸U.⁸⁾ As for non-isotopic members, some volcanoes showed the disequilibria of ²³⁰Th < ²³⁴U and others ²³⁰Th > ²³⁴U. Radioactive disequilibria among U-series nuclides in volcanic rocks will contribute to

volcanology. We are observing activity ratios of $^{230}\text{Th}/^{234}\text{U}$ in rhyolitic volcanic rocks erupted from Izu-Mariana island-arc volcanoes in order to discuss on the radioactive disequilibrium between ^{234}U and ^{230}Th in the late stage of magmatic evolution. The present report deals with the $^{230}\text{Th}/^{238}\text{U}$ activity ratios of 7 rhyolitic products from 2 volcanoes of Izu-Mariana island-arc, Kozu-shima and Niijima, ranged from 0.69 to 0.97, indicating that ^{230}Th was in radioactive disequilibrium with ^{234}U in the erupting magmas.

3P09 : Isotope-geochemical Study on Hot and Mineral Springs around Hakusan Volcano, Japan

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Hot and mineral spring waters, which discharge around Hakusan Volcano located on the boundary between Ishikawa and Gifu Prefecture, were collected and measured for δD , $\delta^{13}\text{C}$, $\delta^{18}\text{O}$ and $\delta^{34}\text{S}$, and concentrations of ^{14}C , U and Ra isotopes, and major dissolved ions. δD and $\delta^{18}\text{O}$ values of samples showed that all of the waters were derived from the local meteoric water. By considering Br/Cl ratios and Li/Na-K/Na relations, saline springs near Hakusan Volcano were suggested to be produced by dissolution of sea salt in the green tuffs. The values of $\delta^{13}\text{C}$ ranged from -6 to -2 ‰ and their ^{14}C concentrations were within the values of 0.24–6.77 pmC. The ^{14}C ages of these waters were tentatively calculated to be 12,000–30,000 years by $\delta^{13}\text{C}$ mixing model. The concentrations of ^{238}U and $^{234}\text{U}/^{238}\text{U}$ activity ratios were observed in the wide range from 0.006 to 14.4 mBq/kg and from 0.89 to 38.4, respectively. The ^{226}Ra concentrations varied from 0.42–242 mBq/kg and the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios from 0.39–38.4. The results will be further discussed including $\delta^{34}\text{S}$.

3P10 : Activity ratio of Ra isotopes leached from monazite

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We are observing the leaching behavior of Ra isotopes into a solution from monazite in order to study about the leaching mechanism of Ra isotopes from mineral. The activity of Ra isotopes in leachate from monazite was observed to decrease with increasing pH of the solution. The leaching of Ra isotopes was dependent on the difference in the history of decay processes of Ra

isotopes which have formed.

3P11 : Vertical distribution of $^{228}\text{Ra}/^{226}\text{Ra}$ ratio and ^{137}Cs concentration in the Sea of Japan

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As radionuclides have their own origin, half-life and geochemical behavior, they have been used as geochemical tracers of oceanography. Especially, the ^{228}Ra ($t_{1/2} = 5.75$ y)/ ^{226}Ra (1600 y) activity ratio of seawater has been used as powerful tracer for the study of water migrations. Since ^{228}Ra concentration in deep water (>1000 m) is very low level, measurement of ^{228}Ra is not easy without using low-background γ -spectrometry. By the use of ultra low-background well type Ge detectors in Ogoya Underground Laboratory, it became possible to detect ^{228}Ra using only 20-l of water sample. In this study, γ -spectrometry has been carried out to seawater samples collected at two off-shore sites around Yamato ridge (3510 m and 2610 m in depth) and 5 coastal areas of Honshu Island (85–485 m in depth) in the Sea of Japan (May–June 2004). From these results, we discuss vertical migrations of water mass in coastal and offshore areas.

3P12 : Seasonal variation of $^{228}\text{Ra}/^{226}\text{Ra}$ ratio of coastal water in Japan Sea

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Circulations of seawater, especially for coastal water, are expected to bring the information about environmental changes and the transports of pollutants by occasional accidents. The half-life of ^{228}Ra determines the ^{228}Ra activity and $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of seawater, which are sensitive tracers for studies of the horizontal and vertical migration of seawater. In order to investigate seasonal variations of the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of the Sea of Japan, we collected coastal water samples in the coastal area along the Sea of Japan (mainly the Noto Peninsula), and measured by low-background gamma-spectrometry. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of water samples collected at the coastal area of the Noto Peninsula exhibited seasonal variation with minimum values in June ($^{228}\text{Ra}/^{226}\text{Ra} = \sim 1$) and maximum values in December (1.5–2.5), which was mainly governed by the change in the ^{228}Ra activity. From these results, we assessed the water circulation of coastal

areas of the Sea of Japan.

3P13 : Measurement of radium in hot-spring waters using solid-phase extraction membrane

Saito, T., Nagai, H., (College of Humanities & Sciences, Nihon Univ.)

An analytical method for radium in environmental water using solid-phase extraction membrane (Empore™ Radium Rad Disk) was investigated. Acidified samples (2 – 4 L), which were aliquoted 0.5 – 2M HNO₃, through an Empore™ Radium Rad Disk by a flow rate of 50 mL/min. Radium fraction was eluted by 30 ml of 0.2M diammonium hydrogen citrate with gently vacuum, and analyzed by a well-type HPGe γ -ray spectrometer for the 911 keV γ -ray from ²²⁸Ac, being in radioactive equilibrium with precursors. Radium recovery was virtually 100 % under the condition of 2 M-HNO₃, 2 L and 0.5M-HNO₃, 4 L samples, respectively. Although Tamagawa hot-spring water was dissolved large amount of solutes because of high acidity condition, radium was collected quantitatively. Thus the analytical method may be able to be applied for almost environmental samples.

3P14 : Distribution of ¹⁰Be and ²³⁰Th concentration of marine core sediment from the North Pacific Ocean

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Concentration of ¹⁰Be and ²³⁰Th for the North Pacific marine sediment collected by R/V Hakuho-Maru during KH00-3(2000) and KH03-1(2003) cruise were investigated. Most of the samples showed constant sedimentation rate which were between 2 to 3 mm/kyr estimated from ex.²³⁰Th concentration. For some samples, however, changes in ¹⁰Be concentration were observed. These samples had similar changes in elementary compositions and ¹⁰Be/⁹Be ratio which imply changes in sedimentary environment.

3P15 : Distribution and behavior of trace elements and ¹³⁷Cs in deposition samples collected at Fukuoka in 1964

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Neutron activation analysis (NAA) and γ -ray spectrometry were applied to the deposition samples collected at Fukuoka in 1964 and 2000. The results obtained in this study are summarized as follows; (1) the ratios of the amounts (g) of monthly depositions dried up and the concentrations ($\mu\text{g}/\text{m}^2$) of chemical elements in the samples in 1964 to those in 2000 were 7 (March) to 180 (July) and 6 (Cl) to 300 (Al), respectively. It indicates that the depositions at Fukuoka in 1964 are extremely different from those in 2000. (2) The concentrations of uranium, radioactivities of ¹³⁷Cs and the U/Th ratios in the samples in 1964 were apparently higher between March and May than those in other months. It seems that the depositions in 1964 reflect the active stratosphere-troposphere air exchange in the springtime. (3) It was suggested that the mechanism of the deposition in 2000 was different from that in 1964 although the peak of the concentrations of uranium in 2000 was observed also between March and May, indicating the intrusion of soil dusts (Kosa) originating from the Asian desert and arid areas.

3P16: Distribution of uranium and thorium isotopes in iron smelting process

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A behavior of uranium and thorium isotopes was investigated in iron smelting process. Therefore, uranium and thorium isotopes in iron ore, coke and slag used an industry were determined by an INAA and an alpha spectrometry. In alpha spectrometry, uranium and thorium isotopes were measured with a SmF₃ coprecipitation method. The recovery yield of uranium and thorium isotopes through chemical procedure in the alpha spectrometry was compared with the results of INAA and was good. It was found that ²³⁸U-²³⁴U and ²³²Th-²²⁸Th were radioactive equilibrium in iron ore and slag. The radioactivity ratio of ²³⁸U to ²³²Th in iron ore, slag and coke was 11.1, 2.6, and 1.4, respectively. This means distribution of uranium and thorium is mainly slag in coke due to uranium and thorium though iron smelting.

3P17 : Optimization of biological processes for the removal of arsenic from groundwater based on X-ray spectroscopic analyses

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XANES analyses were conducted to clarify the mechanisms of arsenic removal by iron bacteria. The valence state of As sorbed to the iron and manganese minerals precipitated on the bacterial surface was +5 when As(III) was added to the live bacteria. Elemental analyses showed that the precipitates contained 14%Fe and 5 % P. When arsenic acid was removed by the bacteria in a batch sorption test, equimolar amount of phosphorus was desorbed, indicative of an ion-exchange between arsenic and phosphoric acid. Biological oxidation of As(III) to As(V) by iron bacteria followed by ion-exchange sorption was probably the mechanisms of As(III) removal.

3P18 : Removal of radioactive contaminants from iron pipes utilizing reversed micelle in supercritical CO₂

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Generation of radioactive wastes becomes a serious problem in nuclear industry. A large number of pipes and valves are used in the nuclear power plants for water cooling system. Radioactive contaminants, such as nickel-substituted ferrite (Ni_xFe_{3-x}O₄), are accumulated and fixed on the inside surfaces of those parts. The contaminated pipes and valves are replaced with new ones during the maintenance period, and then discharged as nuclear wastes. Chemical dissolution of these ferrites could remove the contaminants from the surface of the pipes and valves. This treatment, however, will cause larger amount in volume of secondary waste than that of the original contaminants. For volume reduction of wastes, we propose a new method to remove the radioactive contaminants from the materials using reactive microemulsion of organic acids in supercritical carbon dioxide (SF-CO₂). The organic acid was dispersed into SF-CO₂ by polyoxyethylene nonylphenyl ether, pentadecafluorooctanoic acid or acetic acid at 25 MPa. Decontamination of ferrites on iron pipes was conducted at 323 and 353 K.

3P19 : Adsorption behavior of Alkali metal ions and alkaline earth metal ions on tertiary pyridine resin

Sato, M., Suzuki, T., Fuji, Y., (Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology.)

Separation of Cs and Sr which elements are included in the high level radioactive waste is an important matter. Adsorption and separation of these elements by using tertiary pyridine resin were investigated nitric acid or hydrochloric acid – methanol mixed solvent system. As ratio of methanol increases, the distribution coefficients of Sr and Cs increased in nitric acid system. The separation factor also increased with methanol ratio. We confirmed that Cs and Sr are almost separated in case of nitric acid : methanol = 4:6. On the other hand, the hydrochloric acid / methanol mixed system had a maximal value of distribution coefficients. The separation of Cs and Sr in hydrochloric acid system was insufficient in comparison with nitric acid system.

3P20: Positron annihilation in solid and liquid with a Coincidence Doppler broadening spectroscopy (CDBS)

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A coincidence Doppler broadening spectroscopy (CDBS) is useful to study minute elements in compounds and polymer materials. The spectra can be obtained by the coincidence of two Ge detectors placed at the 180 degree configuration, which can provide an extremely low BG (SN ratio 10⁶ ~ 10⁷) comparing with BG obtained by a single Ge detector. This technique was applied to solid and liquid n-hexane (n-C₆H₁₄, mp: -94°C) In liquid state Ps creates its own bubble and annihilates inside: in solid state, Ps annihilates in intermolecular spaces. At LN2 temperature, the lifetime is about 1.2ns and 4ns at RT. At dry ice temperature, n-hexane is liquid but due to the large surface tension, annihilation in the bubble is similar to the annihilation in solid even though its lifetime is longer than that in solid. In the bubble created at RT, CDBS is distorted at high momentum part, which can be explained by the wall effect of the surface tension.

3P21 : Study on presolar grains using multiparameter coincidence method

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High sensitive trace element analyses without chemical separation were carried out by the combination of neutron activation analysis and the method of multi-parameter coincidence spectroscopy. In the case of neutron activation analysis, measurements of γ -rays from trace elements are strongly interfered by the γ -rays from major elements, e.g., ²⁴Na, ⁵⁶Mn. So usually

chemical separation processes are required to eliminate the major elements for determination of the trace elements. In this study, using neutron activation analysis with multi-parameter coincidence spectrometry method which has been developed at JAERI, a non-destructive, high sensitive determination of trace elements has been realized. The multiple coincidence spectrometry measurements are performed with an array of 19 Ge detectors, named GEMINI-II. About 10 ppt of iridium in geological samples can be determined using this system without chemical separations. In this paper, we attempt to measure trace elements in presolar grains extracted from meteorite. Presolar grains contain the original atoms of the stars in which they were formed. They are thus samples of stars we can study in the laboratory and are providing interesting information about a wide range of astrophysical topics.

3P22 : Factors of variations in serum protein binding rates of radiopharmaceuticals for diagnostic imaging by hemodialysis

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The binding affinity of drugs to serum protein, such as human serum albumin (HSA) or α_1 -acid glycoprotein, gives variations on permeability to the target organ. In case of hemodialysis patients, the rate of serum protein binding of drug is expected to change between pre- and post-hemodialysis. We selected two diagnostic imaging pharmaceuticals for cerebral and cardiac diseases, ¹²³I-N-isopropyl-p-iodoamphetamine (I-IMP) and ^{99m}Tc-tetrofosmin (Tc-TF). The free fraction rates of radiopharmaceuticals in pre-hemodialysis, were 1.15 (Tc-TF) -1.31 (I-IMP) times as higher as in post-hemodialysis. The variations of protein binding rates by hemodialysis were relative to behavior of radiolabeled site-markers, hemoconcentration of dialysis, and changes of endogenous substance concentrations. It was suggested possibility of monitoring for effective administration.

3P23 : Analysis of oxidative stress by selenium deficiency and distribution of trace elements in male and female rats

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GSH concentration, SOD and GSH-Px activity in liver, and

contents of iron (Fe), cobalt (Co), zinc (Zn), and selenium (Se) in the organs (liver, kidney, spleen, brain) were measured and the difference between male and female rats in selenium(Se)-deficiency was discussed. In Se-deficient(SeD) group of male rats, the contents of Fe in the liver, kidney, and spleen increased, while, in female, the contents increased only in the liver, whereas it decreased in the kidney compared with Se-control rats. GSH-Px activity in SeD group of male rats decreased remarkably. SOD activity in SeD group of male rats in liver decreased in comparing with normal and Se-control rats. However, significant difference was not indicated among the female groups of SeD, Se-control, and normal rats. GSH concentration in SeD group of male rats decreased with GSH-Px activity, while, in female it increased. It is suggested that female rats were not scarcely influenced by oxidative stress under Se-deficiency compared with male rats.

3P24 : Effects of iron overload, hyperoxia, and Se-deficiency on the redox status in the rat liver

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The effect of iron-overload and hyperoxia on the spin clearance and hydrogen peroxide level in bile of 8 week-old male Se-deficient rats was investigated by X-band ESR. The signal intensity of nitroxyl spin probe, carbamoyl-PROXYL, in bile of rats decreased with time when carbamoyl-PROXYL was iv injected without previous injection of Fe(II) citrate. However, the signal intensity increased after the initial decay, attained a second peak and then decreased for the all the iron overloaded groups (Normal, SeC, and SeD). H₂O₂ concentration was evaluated by the spin trapping method. The concentration in bile increased to high level under the iron overload and also under hyperoxia. The H₂O₂ level of SeD group was higher than normal and SeC groups under air. The H₂O₂ levels were found no remarkable difference among rat groups under hyperoxia and control group. The nitroxyl decay rate in the bile was smaller for the rats under hyperoxia. In vivo redox status was affected by several conditions, such as Se-deficiency, iron overload, and hyperoxia, complicatedly.

3P25 : The maximum tolerated dose (MTD) of ²²⁷Th-EDTMP and ²²³RaCl₂ for rodents.

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^{227}Th -EDTMP and ^{223}Ra , which are promising agents for treatment of bone metastasis, selectively accumulate in bone and retain for a long time. The decay and growth patterns of radioactivity of ^{227}Th and ^{223}Ra after accumulation in bone are specific for each nuclide. In the case of ^{223}Ra pattern, total radioactivity decays according to ^{223}Ra half-life with 4 α -emissions due to the radioactive equilibrium with its progeny. On the other hand, in the case of ^{227}Th pattern, at first total activity increases with time and reaches the maximum level, and then decays according to ^{227}Th half-life. Therefore, ^{227}Th administration method gives effectively prolongation of α -radiation dose and may gain the maximum tolerated dose (MTD) in comparison to ^{223}Ra administration method. We attempt here to determine the MTD of ^{227}Th -EDTMP and ^{223}Ra for mice and rats. Four-week female Sprague-Dawley rats were injected different dose of ^{227}Th -EDTMP or ^{223}Ra . The blood was collected before injection and several days after injection, and red blood cells, white blood cells, platelet were counted. The effect of half-life of ^{227}Th -EDTMP and ^{223}Ra on MTD will be discussed.

3P26 : Production and application of lead-203 (^{203}Pb) isotope for nuclear medicine

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Several radioisotopes (RI), such as ^{64}Cu , ^{90}Y , ^{213}Bi , and ^{211}Pb , have been using for tumor therapy. However, it is difficult to study the biodistribution of these RI during long period and to evaluate the radiation dose due to those short half-life. Lead-203 has been thought as a suitable RI for alternative tracer of ^{211}Pb . In this study, we made a ^{203}Pb tracer via the $^{203}\text{Tl}(p,n)^{203}\text{Pb}$ reaction and evaluated the use of ^{203}Pb for nuclear medicine. Lead-203 was produced with high yield radioactivity and the separation of ^{203}Pb from target was simple and easy. After radiochemical separation, ^{203}Pb with saline solution was mixed with DOTMP chelate and injected into mice. After appropriate time intervals, the blood, femur, kidney, liver, and spleen were excised and subjected to gamma-ray spectrometry to determine uptake rate (% injected dose per gram). As the results, renal ^{203}Pb uptake was significantly decreased by DOTMP chelation in the comparison with that of $^{203}\text{PbCl}_2$. These trends were similar to the $^{223}\text{Ra}(^{211}\text{Pb})$ injection experiment. Therefore, ^{203}Pb is an alternative tracer for ^{211}Pb .

3P27 : Measurement of hyperfine fields in metal chelate complexes by the PAC method

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We reported on the TDPAC measurement using a ^{117}Cd probe at the metal site of mavidcyanin, which is a protein molecule with a copper site. It is necessary to check if there is dependence of adopted PAC probes. In order to verify the effect, several 4-coordinate complexes as mavidcyanin were subject to the PAC measurement for comparison in this study. The parent nuclei ^{117}Cd and ^{111m}Cd were obtained by irradiating enriched ^{116}CdO and ^{110}CdO reagents, respectively at Kyoto University Research Reactor Institute. The ^{111}In sample was purchased from Nihon Medi-Physics Co. Ltd. The used ligands are oxine, diethyldithiocarbamate (DDC), dithizone, and benzoylphenylhydroxylamine (BPHA). In Table 1 are shown asymmetry parameters, the values of which in parentheses are from the other measurement, and electric field gradient values (EFG) of hyperfine field obtained from the TDPAC spectra. It demonstrates that the mavidcyanin data are fairly larger than those of all the other complexes although some data are found to depend on the chemical species of probe nuclide. The specific nature of protein in metal coordination seems to affect the field.

3P28 : Nanoporosity of SiOCH-based plasma enhanced chemical vapor deposition films studied by variable-energy positron annihilation

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We studied formation of subnanopores in SiOCH-based plasma enhanced chemical vapor deposition (PECVD) thin films by measuring the lifetime of positronium (Ps: the bound state of a positron and an electron) with the variable-energy positron annihilation technique. The films with several hundred nm thicknesses were deposited on silicon wafers in a parallel plate PECVD reactor using a 13.56 MHz rf at a substrate temperature of 300 °C. A mixture of tetraethyl orthosilicate (TEOS) and hexamethyldisiloxane (HMDSiO) with their ratio ranging from 0 to 1 diluted in argon and oxygen was used as a precursor. With increasing the HMDSiO fraction Ps lifetime increases from 2.0 ns to 6.4 ns. This indicates that pores of 0.1 nm³-0.6 nm³ size can be

introduced by the PECVD method.

3P29 : Characterization of silica-gel pores by Doppler broadening of prompt gamma-ray

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We report on an application of the Doppler broadening of prompt gamma-ray to characterization of silica-gel pores. The prompt gamma-rays are emitted from ${}^7\text{Li}$ produced in the ${}^{10}\text{B}(\text{n}, \alpha){}^7\text{Li}$ reaction. Three types of silica gels (A, B, ID type) were used here; the mean pore diameters and specific surface areas were $2.2\text{nm}/650\text{m}^2\text{g}^{-1}$, $7.0\text{nm}/450\text{m}^2\text{g}^{-1}$, and $15.5\text{nm}/310\text{m}^2\text{g}^{-1}$, respectively. The pores of silica gels were just filled with an aqueous solution of boric acid, and then were dried by heating in air. Both samples before and after removing water were submitted to a prompt gamma-ray measurement in the neutron-beam guide of JRR-3M in JAERI. By analyzing the Doppler broadened spectrum, a degradation constant D was evaluated, which is a reciprocal of time constant of ${}^7\text{Li}$ degradation in media. Our previous work showed that D should reflect the elemental composition and the average density of a material where energetic ${}^7\text{Li}$ ions move and lose their kinetic energy. Relation between the D values and properties of silica gel were discussed in the present work.

3P30 : Quadrupole relaxation of ${}^{140}\text{Ce}$ in highly oriented pyrolytic graphite

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The time-differential perturbed angular correlation (TDPAC) technique has been applied to the study of the electronic state and dynamic behavior of a probe, ${}^{140}\text{Ce}$ as a β -decay descendant of a fission product ${}^{140}\text{Cs}$, implanted in highly oriented pyrolytic graphite (HOPG). Temperature-dependent gradual attenuation of the directional anisotropy has been observed for the time spectra, which implies the nuclear spin relaxation caused by a thermally activated dynamic interaction between the probe nuclei and the extranuclear field. This observation is contrastive to the case with TDPAC measurements using another probe, ${}^{19}\text{F}$, introduced in the same sample. In this paper, detailed discussion is made for the

results based on an applied magnetic-field dependence as well as the temperature dependence of the time spectra.

3P31 : Chemical states of ${}^{57}\text{Fe}$ decayed from ${}^{57}\text{Mn}$ after implantation into solid oxygen

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We have performed an in-beam Mössbauer spectroscopic experiment using a short-lived ${}^{57}\text{Mn}$ beam, in order to study the production of the novel chemical species of ${}^{57}\text{Fe}$ atoms arising from ${}^{57}\text{Mn}$ implanted into solid oxygen. ${}^{57}\text{Mn}$ particles were produced as a secondary RI beam following a nuclear projectile-fragmentation reaction of ${}^{58}\text{Fe}$ beams with Be production target, and separated by the in-flight isotope separator using RIPS in RIKEN Accelerator Research Facility. The solid O_2 sample was obtained by an aggregation of a pure O_2 gas in an Al plate cooled at 18K for 4 hr. The in-beam Mössbauer spectra were measured at 18 K during the implantation of ${}^{57}\text{Mn}$. From the basis of Mössbauer parameters of the isomer shifts and the quadrupole splittings and the ab-initio molecular orbital calculations, the 4 components were assigned to be $\text{Fe}(\text{O}_2)$, FeO , $\text{Fe}(\text{O}_2)_2$, and a chemical species in a higher oxidation state of Fe atoms.

3P32 : Mössbauer study of iron oxide films produced by pulsed laser deposition

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Iron oxide films were deposited by pulsed laser ablation of hematite and magnetite. When laser-evaporated atoms were deposited on a substrate of aluminum foil, the temperature of the substrate was kept at desired temperature (10 ~ 573 K) using a closed-cycle helium refrigerator or a resistive heater. Mössbauer spectra of the prepared films were measured at room temperature in transmission or conversion-electron geometry using ${}^{57}\text{Co}/\text{Rh}$ or ${}^{57}\text{Co}/\text{Cr}$ source. Mössbauer spectra of the oxide films deposited on aluminum substrate at 297K and 573K by laser ablation of hematite showed a doublet and two sextets. The doublet absorption is assigned to Fe^{2+} and the two sextet absorptions are assigned to Fe_3O_4 . Oxygen atoms of Fe_2O_3 are released on the deposition process, and a film has less amounts of O-atom than a target

material. At the low temperature (10K), two sextet absorptions disappeared. It is considered to be why the deposited atoms have too small diffusion energy to form Fe_3O_4 crystal.

3P33 : ^{197}Au Mössbauer spectra of cyclometallated gold complexes having arsenic bridging ligand

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The electronic states of cyclometallated gold complexes having arsenic bridging ligand are investigated by ^{197}Au Mössbauer spectroscopy. ^{197}Au Mössbauer isomer shifts (δ) and quadrupole splittings (Δ) reflect the electron donor ability of the ligands sensitively. Both δ and Δ values for the soft ligands such as carbon or arsine are larger than those for hard ligands like halogens. ^{197}Au Mössbauer parameters for the gold(I) complexes having arsine-containing ligands are smaller than those for phosphorous analogue, suggesting the weaker Au-As bond. No difference in ^{197}Au Mössbauer parameters between $[\text{Au}_2\{(\mu\text{-C}_6\text{H}_3\text{-5-Me})\text{AsPh}_2\}_2]$ and $[\text{Au}_2\{(\mu\text{-C}_6\text{H}_3\text{-6-Me})\text{AsPh}_2\}_2]$ are observed, suggesting that the electronic states of the gold atom are not affected by the position of the substituted methyl group.

3P34 : Mössbauer study of forging flake produced in Tataro iron manufacturing process

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A lump of iron was obtained by Tataro iron manufacturing process. Forging flakes scaled off during the forging process of the iron lump. ^{57}Fe Mössbauer spectra of these flakes were taken in order to know which minerals these flakes consisted of. From the results of measurements it was found the forging flake consisted of magnetite, haematite, wustite and fayalite. A small amount of metallic iron was observed in samples produced at the early stage of forging, which indicates that at the early stage of forging the surface of iron lump was fragile and the forging flake was easy to scale off. Fayalite was produced by the reaction between the iron and muddy water used at the forging process.

3P35 : Mössbauer studies of trace ^{57}Fe -doped SnO_2

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Diluted magnetic semiconductors (DMS) have been of great

interest to realize spintronics in near future. SnO_2 doped with 0.5, 1, 3 and 5% ^{57}Fe were prepared by a sol-gel method, and characterized by ^{57}Fe Mössbauer spectrometry, and XRD. The prepared samples were annealed at 500 °C, 600 °C and 650 °C for 2 hrs. The single phase of SnO_2 was recognized in all XRD patterns of prepared SnO_2 powders although the higher annealing temperature provided the better crystallinity. Magnetic relaxation peaks were observed in addition to the doublet of $\text{Sn}_{1-x}\text{Fe}_x\text{O}_2$ solid solution in ^{57}Fe Mössbauer spectra of SnO_2 doped with <5% ^{57}Fe . The amount of the magnetic relaxation components increased with the decrease of the amount of doped ^{57}Fe . Two phases of paramagnetic $\text{Sn}_{1-x}\text{Fe}_x\text{O}_2$ solid solution and anti-ferromagnetic $\alpha\text{-Fe}_2\text{O}_3$ were observed in only Mössbauer spectrum of 5% ^{57}Fe doped SnO_2 , annealed at 650°C. It was found that SnO_2 doped with diluted Fe is one of candidates for DMS materials.

3P36: Measurements of electronic KX rays emitted in the formation process of muonic atoms.

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The formation process of muonic atoms has been studied well, on the other hand, there is still room to examine the electron rearrangement process after the muon capture. We have measured the electronic X rays by using HPGe detectors for various elements to investigate the electron rearrangement, because the electronic X-ray energies are influenced by the electron configuration of the muonic atoms. All measurements were performed at the μ -port of the Muon Science Laboratory at KEK. Electronic KX-ray energies of muonic tin, barium, holmium, tantalum, lead, thorium and uranium were compared with previous works' values (only measured for heavy muonic atoms($Z>72$)).

3P37 : A study of depositional environment of the Antarctic Ocean sediment containing an extraordinary iron species.

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In order to investigate depositional environment of the Antarctic Ocean, we have studied ^{57}Fe Moessbauer spectra of its sediment in detail and measured XANES spectra. We have already reported that

its Moessbauer spectrum contains peculiar doublet. As a result of detail calculation for the spectrum, it was confirmed that this chemical species existed (I.S. 0.567-0.590mm/s, Q.S. 1.701-1.827mm/s, H.W. 0.376-0.468mm/s). This species is not found in past reports. As a result of the XANES measurement, the sediment spectrum that in the passed between hematite and olivine was observed near the Fe K-edge. In addition, the spectrum tended to correspond to the past change of events paleoceanographic slightly. It is predictable that depositional environment of the collection point was relatively anoxic atmosphere from these results, and this species has the possibility of immature glauconite ((K, Na, Ca)(Fe³⁺, Al, Mg, Fe²⁺)₂(Si, Al)₄O₁₀(OH)₂).

3P38 : Focused neutron beam induced prompt gamma-ray analysis at JAERI

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We have installed a neutron focusing beam guide for the thermal neutron induced prompt γ -ray analysis (PGA) system at JRR-3. The guide was designed with multichannel structure, seven channels in this case, to focus a neutron beam of wide area effectively. Focusing neutron beam distribution was measured with a CCD system and it detected that a focal area was 1.5 x 4.8 mm² at a PGA sample position. The focal area flux was measured by the gold tin film activation method and was 5.7 x 10⁸ n.cm⁻².s⁻¹. It was clear that focusing gain in peak area intensity at the focal point was 3.5 in peak area intensity at the focal point. The focused neutrons have been used to enhance the efficacy of the PGA. We measured a small chip of BCR 680 (8.8 mg 2.2 mm ϕ x 2.8mm). The results with and without the focusing beam are compared. It was clear that with focusing beam the signal intensity for Cd 558 keV peak was much higher, with a gain of 3 and an S/N ratio of 2.

3P39 : A study on the distribution of cobalt in *Clethradease* leaves

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Clethra barbinervis is known to accumulate cobalt (Co) in anomalously high contents.¹ In order to elucidate the feature of Co in the leaves of *Clethra*, we have investigated two-dimensional distribution of Co in leaves on an imaging plate (IP). Neutron

emission of leaves non-selectively activated such metals contained in the leaves as ⁶⁰Co, ⁶⁵Zn, ¹³⁴Cs, ⁸⁶Rb, ¹³¹Ba, ⁴⁶Sc, ⁵⁹Fe, ^{115m}Cd, ³²P, and ⁴⁵Ca. In order to achieve selective imaging of Co, we have explored solvent extraction to eliminate possible influence of radiation from the metals but ⁶⁰Co. Treatment of the leaves with sodium bicarbonate eliminated Cs and Rb, but left Co as well as Zn, whose emission is mostly γ -ray. Other metals remained were ¹³¹Ba, ⁴⁶Sc, ⁵⁹Fe, ^{115m}Cd, (³²P). Half-lives of them are short enough to wait for their decay until submission to IP. The β -ray from ⁴⁵Ca was effectively shielded with aluminum foil. Thus, the distribution of Co was successfully detected on IP. [1] N. Yamagata, S. Murata, Kosu Eiseiin Kenkyu Hokoku (1964) 13 (3), 170-5.

3P40: The determination of trace manganese in high-purity iron

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Trace amount of manganese in high-purity iron sample JSS001-4, JSS003-5 and JSS009-2 was estimated by two methods; instrumental neutron activation analysis using Tc-Pn in Research Reactor Institute, Kyoto University and inductively coupled plasma mass spectrometry after separating iron by anion exchange column chromatography, respectively. From the result of both quantitative analysis, we are going to estimate the interference by ⁵⁶Fe(n, p)⁵⁶Mn reaction with fast neutron in INAA using Tc-Pn in KUR. Those method in Tc-Pn in KUR will be applicable to determination of trace amount of manganese in iron meteorites in the near future.

3P41 : Determination of trace elements in hepatocyte of zinc deficient mice

Kamishima, J., Ogi, T., Minayoshi, R., Kinugawa, N., Suganuma, H., Noguchi, M., Ishikawa, K., Yanaga, M., (Fac. of Sci., Shizuoka Univ.)

Eight-week-old male mice of ICR strain were divided into two groups; one was fed with zinc deficient diet, the other with control diet. After 3 weeks of treatment periods, their livers were removed. Two types of experiments were performed. In the first experiment, the liver samples were centrifuged under differential conditions in order to separate into cellular fragments and 5 subcellular fractions, such as nuclear, mitochondrial, lysosomal, microsomal and cytosolic fractions. Each fraction was freeze-dried for INAA. Concentrations of 11 elements were determined by INAA. In the

second experiment, SDS-PAGE was performed for cytosolic fraction. Zinc concentration in cytosolic fraction of zinc deficient mice was lower than that of control one. On the other hand, cobalt concentrations in all fractions of zinc deficient mice were higher than those of control ones. However, when the positions and the number of the protein bands on gel developed for the cytosolic fraction of zinc deficient mice were compared with those of control ones, no significant differences were found between them.

3P42 : Change of concentrations of trace elements in livers of zinc deficiency mice (2)

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Concentrations of trace elements in livers of Zn deficient mice and control mice were investigated. Eight-week old male mice of ICR stain were divided into two groups. One group was fed with Zn-deficient diet, and the other group with control diet. After three weeks, the livers of each group were removed and homogenized. Then, the supernatant fraction of hepatic cells of each mouse was separated, by ultracentrifugation. The supernatant fraction was further divided into forty fractions by means of gel filtration chromatography. Then, ICP-MS, BCA protein assay method, and SDS-PAGE were carried out for each fraction. The concentrations of zinc were decreased in the 14-17th fractions of Zn-deficient mice, and of cobalt were increased in the 14, 17, and 18th fractions. The concentrations of proteins in these fractions were decreased. However, no significant differences were found on gel after SDS-PAGE for the fractions, No. 12-21, of Zn-deficient and control mice.

3P43 : Extractable Organohalogenes (EOX) and Man-Made Organochlorine Compounds in Masked Palm Civet (*Paguma larvata*)

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A concern about worldwide dispersion of anthropogenic halogenated organohalogenes has grown and resulted in the assessment of these compounds in the environment. Gas chromatograph – mass spectrometry is one of suitable techniques for the qualitative and quantitative analysis for individual compounds. On the other hand, neutron activation analysis can give

a complete picture of the total organohalogenes present in the environmental samples. We attempted to investigate the organohalogen concentrations [extractable organobromine (EOBr) and extractable organoiodine (EOI)] in fat tissue samples of masked palm civet collected in Kamakura, Kanagawa. The results show that the concentrations of EOBr and EOI were 0.11-5.1 µg/g and 0.056-0.13 µg/g, respectively. These values were relatively lower than those in marine mammals. The ratios, EOBr/EOI varied among samples. This result implied that this animal might be contaminated with organobromine compounds produced artificially.

