English Abstracts

SPECIAL LECTURES

1S01: Ion beam solid interactions and application to nano technology

Imanishi, N. (Department of Nuclear Engineering, Kyoto University)

The talk is composed of two topics; one is on the electronic sputtering process and the other is on the nanocrystal fabrication by ion implantation. So far, the electronic sputtering process is still one of open problems especially for tightly bound chemical compounds. The aim of the study is to reveal the dynamic mechanism of secondary ion emission in a MeV energy range by relating it to characteristic physical properties of target materials. We have obtained some interesting results on yields and emission energies of secondary ions emitted from several conductive, semiconductive and insulating solid targets. On the other hand, the purpose of the second study is to fabricate nanocrystals embedded in SiO_2 layers for applying silicon to photo devises. Hydrogen plays an important role in increasing photoluminescence efficiency. Therefore, we have studied the hydrogen termination of silicon dangling bonds on embedded silicon nanocrystals/SiO_2 interfaces, and it was found that in order to increase the efficiency, that is, to trap the implanted hydrogen on the interface of silicon nanocrystal/SiO_2, the silicon implantation dose should be less than about $3x10^{17}/cm^2$.

1S02: Basic study on accelerator-driven subcritical reactor at the Research Reactor Institute, Kyoto University

Mishima, K. (Research Reactor Inst., Kyoto Univ.)

In FY2002, a five year research project on accelerator-driven subcritical reactor (ADSR) system launched in the Research Reactor Institute, Kyoto University (KURRI), with the support of the Technology Development Program for Innovative Nuclear Energy System sponsored by the Ministry of Education, Culture, Sport, Science and Technology (MEXT). In this project, a fixed field alternating gradient (FFAG) synchrotron will be developed and connected to the Kyoto University Critical Assembly (KUCA) to investigate basic neutronic characteristics of ADSR. The accelerator system consists of an ion source, an ion beta, a booster (FFAG) and a main ring (FFAG), generating 150 MeV proton beam with the mean current of 1 μA and the frequency of 120Hz. In the critical experiment, incident energy-dependent multiplication of neutrons will be studied and the experimental results will be analyzed by using Monte Carlo calculation codes MCNP-X and MVP. This project will be a starting point to realize the future plan of KURRI to serve as a center for promoting researches in various fields such as neutron, material, life and medical sciences based on nuclear energy science.

2S02: KEK to Kamioka long baseline neutrino oscillation experiment K2K and future experiment

Nishikawa, K. (Grad. School Sci., Kyoto Univ.)

The collaboration has established the principles of the long baseline neutrino experiment. The data prove the existence of neutrino oscillation with better than 99% confidence level and the indication of the neutrino spectrum distortion is observed. The results of oscillation analysis show that the parameters are consistent with the atmospheric neutrino results from Super-Kamiokande experiment.

3S01: Application of AMS-^{14}C dating for the problems of archaeological and historical sciences

Imamura, M. (National Museum of Japanese History, Japan)

Radiocarbon measurements at 0.3-0.5% precision for 0.5-1.0 mg-sized carbon samples are currently performed in many of AMS (Accelerator Mass Spectrometry) facilities in the world. The recent advancement of AMS-^{14}C precision as well as the availability in exquisite universal radiocarbon calibration database such as INTCAL98 provides us with much better opportunities in its application to researches in archaeology and history, which require precise and accurate ages in the absolute scale. Examples will be given such as those on detailed chronologies of the prehistoric ages of Japanese history, and on precision dating method for historical materials research, which have been performed recently at the National Museum of Japanese History. In particular, the new dates obtained for the beginning of the Yayoi period are discussed with emphasis on their implications to the development of paddy rice cultivation in the Japanese archipelago, and its influence from the East Asian history. The related issues and perspectives will be discussed.

INVITED LECTURE

2S01: Mössbauer studies of $Fe_{1-x}Cu_xCr_2S_4$ chalcogenides with properties of colossal magneto-resistance

Vértes, A. (Hungarian Academy of Sciences & Eötvös Loránd University, Hungary)

Klencsár, Z.[1], Németh, Z.[2], Kuzmann, E.[1], Homonnay, Z.[2], Vértes, A.[1,2], Gritzner, G.[3], Kühberger, M.[3] ([1]Hungarian Academy of Sciences, [2]Eötvös Loránd University, Hungary, [3]Johannes Kepler University, Austria)

The discovery of colossal magnetoresistance (CMR) in manganese-based perovskites has stimulated intense research on the physical bases of the CMR effect [1]. The CMR property of manganites seemed to be explainable by the phenomenon of double exchange [2] and spin disorder accompanying the ferromagnetic metal to paramagnetic insulator transition. Barman *et al.* observed strong negative magnetoresistance above and below Curie temperature in iron doped $La_{0.8}Sr_{0.2}Fe_xCo_{1-x}O_{3-\delta}$ [3]. Surprisingly, the amplitude of magnetoresistance found in these perovskites increases monotonically up to room temperature. Furthermore, the amplitude of negative magnetoresistance in these oxides was found to depend strongly on the concentration of iron in the system. Namely, above $T \approx 150$ K magnetoresistance was found to increase with decreasing iron concentration. At the same time, the iron free $La_{0.8}Sr_{0.2}CoO_{3-\delta}$ perovskite shows negative magnetoresistance effect only around the Curie-temperature. However, the puzzling existence of a considerable intrinsic magnetoresistance reported in $FeCr_2S_4$ chalcogenide spinels [4] - that do not possess manganese, oxygen, perovskite structure, or even a metal-to-insulator transition – indicates that phenomena other than the double exchange effect should also be considered in the explanation of the effect of magnetoresistance observed in these materials. The lecture will give a review on the mentioned results.

[1] R. Helmolt, J. Wecker, B. Holzapfel, L. Schultz, K. Samwer, Phys. Rev. Lett. 71 (1993), 2331-2333.

[2] C. Zener, Phys. Rev. 82 (1951), 403-405.

[3] A. Barman, M. Ghosh, S. Biswas, S.K. De, S. Chatterjee, Appl. Phys. Lett. **71** (1997) 3150.

[4] A.P. Ramirez, R.J. Cava, J. Krajewski, Nature 386 (1997), 156-159.

AWARD LECTURE

3S02: Development of a new TDPAC method —a unique approach to condensed matter physics and chemistry of fullerenes

Sato, W. (Grad. School Sci., Osaka Univ.)

The time-differential perturbed angular correlation (TDPAC) method has long been applied in the field of studies on condensed

matter physics, and has made significant contribution in elucidating the microscopic nature of matter through hyperfine interactions. For opening up a new aspect in this field, unique probes and a method have been developed and applied to the study of solid state physics and chemistry of carbon allotropes in a series of my work. At first, the dynamic and electronic properties of endohedral Ce fullerenes ($Ce@C_{82}$, $CeLa@C_{80}$, $Ce@C_{80}$) were investigated employing the encapsulated ^{140}Ce nucleus as the probe. The followings were plausibly suggested by the observation: (1) the fullerene molecules have a thermally activated rotational motion, each of which has freezing temperature of their own and (2) the encaged Ce atoms take the trivalent state. Secondly, a new probe ^{19}F and an on-line measurement system combined with the ion implantation method have been developed, so that even nuclides having short-lived parents would be applicable to the TDPAC method. As the first application of the system, fullerene C_{60} and highly-oriented pyrolytic graphite (HOPG) were adopted. For the C_{60} sample, fluctuation of the extranuclear field was observed, implying a dynamic motion of the probe nucleus. On the other hand, it has been found that a static perturbation acts at the site of the probe nucleus introduced in the HOPG. Each of the results reflecting the nature of the samples shows the applicability of this on-line method and wider application are strongly expected.

ORAL PRESENTATIONS

1A01: Production of Th-229m in nuclear reactions and its identification by alpha-spectrometry

Kikunaga, H.,[1] Kasamatsu, Y.,[2] Mitsugashira, T.,[3] Takamiya, K.,[4] Ohtsuki, T.,[5] Yuki, H.,[5] Hara, M.,[3] Takahashi, N.,[2] Shinohara, A.[2] Shibata, S.,[4] Hashimoto, T.,[1] Kinoshita, N.,[1] Shibusawa, S.,[6] Yokoyama, A.,[6] Nakanishi, T.,[6] ([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]Grad. School Sci., Osaka Univ., [3]The Oarai-branch, Inst. Materials Res., Tohoku Univ., [4]Res. Reactor Inst., Kyoto Univ., [5]Lab. Nucl. Sci., Grad. School Sci., Tohoku Univ. [6]Faculty Sci., Kanazawa Univ.,)

In order to perform an experimental investigation on the decay property of Th-229m corresponding to an ultra-low energy level of Th-229, we have attempted to produce the atoms of Th-229m in nuclear reactions such as Th-232(gamma, p2n)Ac-229, disintegrating to Th-229, and Th-232(p, p3n)Th-229. The reaction products above were isolated from irradiated targets by using several chemical separation methods and identified by alpha-spectrometry. The alpha radioactivity of the isolated sample of the former was found to grow in the energy region of alpha particles from the decay of Th-229g. A similar growth in activity was also confirmed in the latter. The results demonstrate evidence of the production of Th-229m.

1A02: Cation-exchange behavior and the ionic radius of No^{3+}

Toyoshima, A.,[a,b] Tsukada, K.,[b] Akiyama, K.,[b] Asai, M.,[b] Nishinaka, I.,[b] Kaneko, T.,[b] Ichikawa, S.,[b] Nagame, Y.,[b] Haba, H.,[c] Ishizu, H.,[d] Saito, J.,[d] Tani, Y.,[a] Hasegawa, H.,[a] Sato, W.,[a] Shinohara, A.[a] ([a]Graduate School of Sci., Osaka Univ., [b]Advanced Sci. Res. Center, Japan Atomic Energy Res. Inst., [c]Cyclotron Center, RIKEN, [d]Fac. of Sci., Niigata Univ.)

Although the ionic radii of Md^{3+} and Lr^{3+} were determined by a cation-exchange method, there are no available data on the ionic radius of No^{3+} due to the difficulty of the No^{3+} elution after and/or during the strong oxidation of stable No^{2+}. The purpose of the present study is to determine the ionic radius of No^{3+} by a cation-exchange method. ^{255}No produced in the $^{248}Cm(^{12}C, 5n)$ reaction was transported and collected at the chemistry apparatus. ^{255}No was then dissolved in 0.1 M α-HIB solution and passed through PbO_2 and cation-exchange columns successively. Six molar HNO_3 was finally fed to the cation-exchange column to wash the cation-exchange resin. ^{255}No was detected only in the 6 M HNO_3 fractions but not in 0.1 M α-HIB fractions. It was found that No^{2+} was not oxidized to No^{3+} by the PbO_2 oxidant.

1A03: Nuclear structure of ^{257}No studied by measuring internal conversion electrons

Asai, M.,[a] Tsukada, K.,[a] Ichikawa, S.,[a] Nagame, Y.,[a] Ishii, T.,[a] Nishinaka, I.,[a] Akiyama, K.,[a] Osa, A.,[a] Sakama, M.,[b] Oura, Y.,[c] Sueki, K.,[d] Shibata, M.,[e] ([a]Advanced Sci. Res. Center, JAERI; [b]Dep. Radiologic Sci. and Engineering, Univ. Tokushima; [c]Dep. Chemistry, Tokyo Metropolitan Univ.; [d]Dep. Chemistry, Tsukuba Univ.; [e]Facility for Nucl. Materials, Nagoya Univ.)

Internal conversion electrons following the alpha decay of ^{257}No have been measured by means of alpha-electron coincidence spectroscopy. The nucleus ^{257}No was produced in the $^{248}Cm(^{13}C, 4n)$ reaction and mass-separated by a gas-jet coupled on-line isotope separator. Energies and spin-parities of excited states in the daughter nucleus ^{253}Fm as well as the ground state of ^{257}No have been determined experimentally. The 3/2[622] configuration has been assigned to the 125 keV level in ^{253}Fm and the ground state of ^{257}No.

1A04: Anion-exchange behavior of Db in HF

Tsukada, K.,[a] Haba, H.,[b] Asai, M.,[a] Toyoshima A.,[a,c] Akiyama, K.,[a] Nishinaka, I.,[a] Hirata, M.,[a] Hashimoto, K.,[a] Ichikawa, S.,[a] Nagame, Y.,[a] Nakahara, H.,[a,d] Yasuda, K.,[a] Miyamoto, Y.,[a] Saito, J.,[e] Ito, M.,[e] Ishizu, H.,[e] Goto, S.,[e] Kudo, H.,[e] Tani, Y.,[c] Hasegawa, H.,[c] Sato, W.,[c] Shinohara, A.,[c] Sueki, K.,[f] Oura, Y.,[d] Kinoshita, N.,[g] Kikunaga, H.,[g] Yokoyama, A.[g] ([a]JAERI, [b]RIKEN, [c]Osaka Univ., [d]Tokyo Metro. Univ., [e]Niigata Univ., [f]Univ. of Tsukuba, [g]Kanazawa Univ.)

Fluoride complexing strength of Db and its group-5 homologues, Nb and Ta, and pseudo-homologue Pa on the anion-exchange resin was investigated. The sorption sequence of Ta≥Nb>>Pa in 2-15 M HF was observed in the batch experiment. ^{262}Db and ^{169}Ta were produced in the $^{248}Cm(^{19}F, 5n)$ and $^{nat}Gd(^{19}F, xn)$ reactions, respectively, at the JAERI tandem accelerator. 1702 anion-exchange separations were conducted at 14M HF using the Automated Ion exchange separation apparatus coupled with the Detection system for Alpha spectroscopy (AIDA) with a small size of the column of ⌀1.0 mm i.d. × 3.5 mm. It was found that the fluoride complexing strength of Db is smaller than that of Ta, suggesting that the sequence is Ta=Nb>Db≥Pa at 14 M HF.

1A05: X-ray absorption fine structure spectroscopy of Zr and Hf in HCl solution for chemical characterization of rutherfordium

Haba, H.,[a] Akiyama, K.,[b] Tsukada, K.,[b] Asai, M.,[b] Toyoshima, A.,[b,c] Yaita, T.,[b] Hirata, M.,[b] Sueki, K.,[d] Nagame, Y.[b] ([a]Cyclotron Center, RIKEN, [b]Advanced Sci. Res. Center, Japan Atomic Energy Res. Inst., [c]Graduate School of Sci., Osaka Univ., [d]Dep. of Chemistry, Univ. of Tsukuba)

Previously, we reported anion-exchange behavior of element 104, rutherfordium (Rf), together with the group-4 elements Zr and Hf in 4.0–11.5 M HCl solutions. The structural information of Zr and Hf complexes in HCl are essential to discuss the anion-exchange results of Rf and to perform the relativistic molecular orbital calculations. In the present work, we have measured the Extended X-ray Absorption Fine Structure (EXAFS) spectra of Zr and Hf complexes systematically in 1.0–11.5 M HCl and determined the structural parameters such as atomic number and the number of neighboring atoms and their distance from the central atom. The chloride complexation and hydrolysis of Rf as well as Zr and Hf are discussed based on the change in the complex structure with HCl concentration and also on our anion-exchange results.

1A06: Equilibrium charge state of heavy recoil atom in a helium gas

Kaji, D.,[a, b, c] Morita, K.,[a] Morimoto,[a] K., Goto, S.,[b] Haba, H.,[a] Ideguchi, E.,[a] Kanungo, R.,[a] Katori, K.,[a] Koura, H.,[a, d] Kudo, H.,[b] Ohnishi, T.,[a] Ozawa, A.,[a] Suda, T.,[a] Sueki, K.,[e] Tanihata., I.,[f] Tokanai, F.,[g] Xu, H.,[h] Yeremin, A. V.,[a] Zhao, Y. -L.,[a, i] Zheng, T.[a, j] ([a]RIKEN, [b]Niigata Univ., [c]Univ. of Tokyo, [d]Waseda Univ., [e]Tsukuba Univ., [f]Yamagata Univ., [g]IMP, [h]FLNR, [i]IHEP, [j]Peking Univ.)

In RIKEN, we plan to search for new elements, whose atomic numbers are greater than 113, using a gas-filled recoil separator (GARIS). In order to set the proper magnetic field of the separator for such heavy elements, the knowledge of equilibrium charge state (q-bar) of them in a helium gas is essential. We measured q-bar values of heavy elements from Z = 84 to 111 in order to estimate those with greater atomic numbers. An empirical formula was deduced from these data. We could estimate the q-bar values for atoms with Z≧113 using the formula.

1A07: Energy shifts of electronic X rays from pionic atoms

Ninomiya, K., Kasamatsu, Y., Kikunaga, H., Shigekawa, M., Kinoshita, N., Tani, Y., Hasegawa, H., Yatsukawa, M., Sato, W., Takamiya, K., Matsumura, H., Yokoyama, A., Sueki, K., Hamajima, Y., Miura, T., Shinohara, A. (Grad. School Sci., Osaka Univ., Grad. School Natural Sci., Kanazawa Univ., Res. Reactor Inst., Kyoto Univ., Res. Center Radiation, KEK, Faculity Sci., Kanazawa Univ., Dept. Chem., Univ. Tsukuba, LLRL, Inst. Nature and Env. Technol., Kanazawa Univ.)

Negative pions are captured in matter, resulting in the formation of pionic atoms. Owing to the strong screening effect by the pions, the energies of electronic X rays of pionic atoms shift low compared with the characteristic X-ray energies of the target atoms (atomic number Z; more specifically, they come close to those of Z-1 atoms. We measured electronic X rays from pionic atoms (Zn, Mo, Ag, Sn, Ta and (Pb)) precisely at the pi-mu-channel of KEK-PS. We examined the relation between the energy shift from Z-1 atoms and the atomic number Z.

1A08: *c*-Axis orientation dependence of TDPAC for a single-crystalline sample

Sato, W.,[a,b] Ueno, H.,[a] Watanabe, H.,[a] Miyoshi, H.,[c] Yoshimi, A.,[a] Kameda, D.,[c] Kaihara, J.,[c] Shimada, K.,[c] Ito, T.,[c] Suda, S.,[c] Kobayashi, Y.,[a] Asahi, K.[a,c] Shinohara, A.[b] ([a]RIKEN, [b]Grad. School Sci., Osaka Univ., [c]Dept. Phys., Tokyo Inst. Tech.)

The on-line time-differential perturbed-angular-correlation (TDPAC) method using a probe of ^{19}F was applied to the investigation of the orientation of the electric field gradient (EFG) acting at the site of the probe nucleus in an axially symmetric highly-oriented pyrolytic graphite (HOPG). As theoretically predicted, the TDPACs observed with BaF_2 scintillators placed in such different directions that they are perpendicular and parallel to the *c*-axis of the HOPG have shown distinct perturbation patterns from each other. From the analysis using time-differential perturbation factors for which the directions of the principal axis of the EFG are assumed, it has been found that the EFG is oriented along the *c*-axis of the HOPG. The magnitude of the EFG was optimized by least-squares fits of the TDPAC spectra, resulting in good agreements with literature values. These results strongly suggest the applicability of this on-line method to the study of single-crystalline samples.

1A09: HPLC elution behavior of actinium metallofullerenes

Akiyama, K.,[a] Sueki, K.,[b] Tsukada,[a] K., Haba, H.,[c] Asai, M.,[a] Kikuchi, K.,[d] Nagame, Y.,[a] Kadata, M.,[d] Nakahara, H.[d] ([a]Advanced Sci. Res. Center, JAERI, [b]Dept. Chem., Univ. Tsukuba, [c]Cyclotron Center, RIKEN, [d]Dept. Chem., Tokyo Metro. Univ.)

HPLC elution behavior of the actinium metallofullerenes on the Buckyclutsher column (tri-2, 4-dinitrophenyloxa stationary phase) was reported. This stationary phase can separate fullerenes and metallofullerenes from each other by the degree of the polarity caused by such as the distortion of the cage shape and the intra molecular charge transfer from the inside metal atom to the outside fullerene cage and is usually employed for the separation of the fullerene structural isomers. We injected the crudely separated $Ac@C_{82}$ to this stationary phase and found at least two dominant elution peaks in the HPLC elution curve. This result suggests the existence of the plural structural isomers for the $Ac@C_{82}$. It is interesting that $Ac@C_{82}$ has plural isomers in spite of no isomers for the $La@C_{82}$, which forms the same type of $M@C_{82}$ with Ac metallofullerenes.

1A10: Adsorption behavior of trivalent actinides and lanthanides on anion exchange resins in hydrochloric acid / alcohol mixed solvents

Ikeda, A.[a], Suzuki, T.[a], Aida, M.[a], Fujii, Y.[a], Itoh, K.[b], Hara, M.[c], Mitsugashira, T.[c], Ozawa, M.[a,d] ([a]Res. Lab. Nucl. Reactors, Tokyo Inst. Technol., [b]Graduate School of Material Sci. Eng., Shibaura Inst. Technol., [c]IMR, Tohoku Univ., [d]JNC)

The adsorption behavior of trivalent actinides (Am, Cm and Cf) and lanthanides were examined by using several anion exchange resins in hydrochloric acid (HCl) / alcohol mixed solvents. The trivalent actinides were adsorbed far more strongly on tertiary pyridine resins than lanthanides in 70 vol%-conc. HCl / 30 vol%-methanol mixed solvent and were separated from lanthanides. On the other hand, no adsorption and separation phenomena were observed by using typical strongly basic anion exchange resin Dowex 1X8 (quaternary ammonium type) in the same mixed solvent. Therefore, tertiary pyridine resins have a special effect on the separation of trivalent actinides from lanthanides in alcoholic HCl solutions. The distribution coefficients (K_d) of trivalent actinides and lanthanides on tertiary pyridine resins increased with the increase of methanol in mixed solvents.

1A11: Temperature effect on extraction of Eu(III) and Am(III) by TODGA

Arisaka, M., Kimura, T. (JAERI)

Temperature effect on extraction of Eu(III) and Am(III) by N,N,N',N'-tetraoctyl-3-oxapentane- 1,5-diamide (TODGA) was studied in order to evaluate the performance of TODGA as an extractant. Enthalpy (ΔH) was calculated using the van't Hoff equation, and free energy (ΔG) and entropy (ΔS) for the extraction reaction were calculated using the log K_{ex} and ΔH values at 298 K. Thermodynamic parameters for Eu(III) were estimated to be ΔG = −38.2 kJ/mol, ΔH = −120.2 kJ/mol and ΔS = −275.2 J/mol·K. The extraction of Eu(III) with TODGA was more exothermic than that with CMPO (ΔH = −64.5 kJ/mol). This indicates that TODGA is an efficient extractant in comparison with CMPO. In addition, from the measurements of luminescence lifetime of extracted Eu(III), it was found that the water molecules bound to Eu(III) directly were fully replaced by TODGA and/or NO_3^- throughout the extraction.

1B01: Determination of trace elements in iron and steel by neutron activation analysis with multiple gamma-ray detection

Kimura, A.,[a] Toh, Y.,[a] Oshima, M.,[a] Hatsukawa, Y.,[a] Goto, J.,[a] ([a]JAERI)

In recent years, the promotion of recycling of steels is desired from the viewpoints of effective usage of resources and restraint of CO_2 emission. On the other hand, characteristics of recycled steel are hardly affected by some trace elements. For that reason, new techniques to quantitate the trace elements in steel are demanded.In this report, trace elements, As and Sb, in iron certified reference materials were determined by neutron activation analysis with multiple gamma-ray detection method. For determination of As, two samples (JSS001-5, JSS168-7) were irradiated for 10 minutes at

neutron flux of $5.2 \times 10^{17} m^{-2}s^{-1}$ and measured by multiple gamma-ray detector GEMINI-II ,which consists of 16 Ge detectors with BGO Compton suppressor, for 12 hours. For determination of Sb, seven samples (JSS001-5, JSS003-2,4 and so on.) were irradiated for 5 hours at neutron flux of $9.6 \times 10^{17} m^{-2}s^{-1}$ and measured by GEMINI-II for 24 hours. The determined values were in good agreement with the certified and reference values. The lower limit of determination values for As and Sb in high purity iron were 0.01 and 0.003ppm.

1B02: Ultra-trace determination of iridium in geological samples by means of multiparameter coincidence method

Hatsukawa Y., Toh Y., Kimura A., Hosein G., Oshima M., Toyota K., Noguchi T. (JAERI)

In this study, feasibility of the method of the multiparameter coincidence γ-ray spectrometry for neutron activation analysis was apply for ultra-trace determination of iridium in geological samples. In the case of neutron activation analysis of iridium, measurements of γ-rays from Ir are strongly interfered by the γ-rays from major elements, e.g., ^{46}Sc, ^{51}Cr. So usually chemical separation processes are required to eliminate the major elements for determination of the trace elements. Gamma-gamma coincidence of multiple γ-rays from the radioisotopes produced by neutron capture reactions was measured with an array of 12-Ge detectors with BGO Compton suppressors, GEMINI, located at the tandem accelerator facility in JAERI. 10 ppt of iridium in rock sample was determined without chemical separation using multiparameter coincidence spectrometry. Iridium determination in Cuban sediment samples and sediments in deep sea will be discussed.

1B03: Determination of lanthanoids, thorium and uranium in Tokyo Bay marine sediments by neutron activation analysis and behaviors of their elements

Honda, T.,[a] Kawamura, S.[b] ([a]Atom. Ener. Res. Lab., Musashi Inst. Tech., [b]Fac. Eng., Musashi Inst. Tech.)

The neutron activation analysis (NAA) was applied to the marine sediment core samples collected at four locations at Tokyo Bay. The results obtained in this study are summarized as follows: (1) Eleven lanthanoids (the maximum), thorium and uranium were determined by the NAA incorporating the pretreatment using the ultrasonic wave. (2) Lanthanoids abundance patterns show the negative slopes in all core samples as the terrigenous material, i.e. rock and the larger grain size samples, the smaller slopes. In addition, the smaller grain size samples from the mouth at Tokyo Bay revealed the positive Ce anomaly between La and Ce, indicating that the samples from there had the complicated composition. (3) As regards the vertical profiles of the Th and U concentrations, they show the change in the range of several times to a figure of magnitude in some samples. They also show the symmetrical distributions near the surface layers, suggesting that the chemical behaviors of Th and U are different from each other.

1B04: Neutron activation analysis of suspended particulate matters collected at fifteen areas in Aichi Prefecture

Suzuki,K.,[1] Honda,T.[2] ([1]Res. Div. in Eng., Musashi Inst. Tech., [2]Atom. Ener. Res. Lab., Musashi Inst. Tech.)

Neutron activation analysis of suspended particulate matters (SPM) collected at fifteen areas in Aichi Prefecture in summer and winter was done. The Andersen low volume air sampler was used for the sample collection. According to the F value it could be confirmed that Cr, Al, Sc and Ti were the crust origin elements because of a lot of distributions over larger particles (PM2.1<), whereas Br, Zn and V were the artificial origin elements because of the distributions over smaller particles (PM2.1>). As regards the enrichment factor, it tended that the ratios of the abundance of the elements in the SPM to the crust using Al as an indicator were low in the crust origin elements, whereas high in the artificial origin elements. The ratios of the concentrations of the elements at each area to the average at fifteen areas were smaller than others regardless of the season or the particle size, indicating the feature of suburbs.

1B05: Neutron activation analysis of ultra trace Scandium in natural water

Nomura, M., Oura, Y., Amakawa, H., Ebihara, M. (Faculty of Sci.,Tokyo Metropolitan Univ.)

We developed a new method for the determination of ultra trace Sc in natural water sample by instrumental neutron activation analysis (INAA). In this method, a carrier-free ^{47}Sc was spiked to the seawater and Sc was co-precipitated with Fe as hydroxides. The precipitate was separated from solution by centrifugation and then dissolved in 6M HCl. The solution was passed through an anion exchange column to remove Fe. The effluent was evaporated and finally dried on a filter. Subsequently the filter was sandwiched in polyethylene sheets to determine the recovery of Sc by measuring the activity of ^{47}Sc, which ranged from 87 to 97%. Then, the filter was irradiated with neutron for 20 min at JRR-4 reactor. After the irradiation, Sc (^{45}Sc) abundance was determined by measuring ^{46}Sc in gamma-ray spectrometry. The blank corrected Sc concentrations (0.33 ± 0.06ng/kg) were consistent with literature values. It is expected that the accuracy of the present data will be improved by using high purity polyethylene sheet, considering that most of the blank was contributed to the polyethylene sheet used.

1B06: Determination of toxic trace elements in soils and sediments of Bangladesh using Instrumental Neutron Activation Analysis (INAA)

Latif, Sk.A.[1], Halim, M.A.[1], Uddin, M.S.[1], Naher, K.[1], Islam, M.N.[1], Ahmed, F.U.[1], Islam, M.A.[2], Afroj, D.[3], Oura, Y.[3], Ebihara, M.[3], Katada, M.[3] ([1]INST, Bangladesh Atomic Energy Commission, [2]Dept. of Physics, BUET, Bangladesh, [3]Graduate School of Science, Tokyo Metropolitan Univ.)

Pollution studies are essential to an understanding of toxicity of the element in the environment. In Bangladesh the environmental pollution problem has been becoming worst day-by-day. Bangladesh is now facing a serious problem about arsenic (As) and chromium (Cr), which contaminate our environment. To assess the potential sources particularly of As, a parallel study of groundwater, surface water, sediments and soils have been conducted. Arsenic and chromium concentration measurements in soils and sediments were carried out using instrumental neutron activation analysis at JRR-4. The irradiations were performed using TB pipe for 20 min. To evaluate this method IAEA SRM soil-5 was simultaneously irradiated. The results are in good agreement (+/-10%) with the certified values. The soil samples collected from different parts of Bangladesh were highly contaminated by As. In this study, an excessive amount of chromium was determined in sediments of tannery industries areas of Bangladesh.

1B07: Vertical distribution of elements in estuarine sediments

Takahashi, O., Kuno, A., [a]Sugimori, K., Matsuo, M. (Graduate School of Arts and Sci., The Univ. of Tokyo, [a]Dept. of Biology, Toho Univ. School of Medicine)

We collected sediments vertically in Isahaya-bay, Sanbanse-tideland and Sakai-river. These sediments were cut in every 3cm length. Using Instrumental Neutron Activation Analysis and Prompt Gamma-Ray Analysis, we got vertical distribution of over thirty elements. Chemical states of iron in the sediments were investigated by using Moessbauer spectroscopy. Some elements have certain tendency against their depth. By calculating correlation coefficient, some elements were shown to have correlation with each other. For example, I, Br and Cl had

positive correlation. On the other hand, H had negative correlation with Si. In Isahaya-bay sediments, H became rich and Si became poor in deeper sediments, while in Sakai-river sediments, the tendency against depth was opposite, H became poor and Si became rich in deeper sediments. The negative correlation shows possibility of certain chemical reactions. It is assumed that this negative correlation fit to the result of early diagenesis.

1B08: Study on residence time and removal of atmospheric aerosol by use of natural radionuclides

Nishio, S.,[a] Momoshima, N.[b] ([a]Graduate school of Science and Technology, Kumamoto University. [b]Faculty of Science, Kumamoto University)

Radionuclides in the atmosphere are adhered to atmospheric aerosol. Residence time of atmospheric aerosol can be estimated by activity ratio $^{210}Bi/^{210}Pb$ or $^{210}Po/^{210}Pb$ under assuming all activities of ^{210}Pb, ^{210}Bi and ^{210}Po are derived from decay of inert gas ^{222}Rn. To make clear the behavior of atmospheric aerosol, ^{210}Po activity collected on a filter was measured periodically and the initial activity ratio $^{210}Po/^{210}Pb$ is estimated by a ^{210}Po activity growth curve. For the 18 samples collected from 18 November 2002 to 22 May 2003, the initial activity ratios $^{210}Po/^{210}Pb$ and the residence times were in the range of 0.05-0.22 and 7-40 days, respectively. Atmospheric aerosol is effectively removed from atmosphere by precipitation, but the activity ratio $^{210}Po/^{210}Pb$ was not correlated with precipitation of the sampling period. The activity ratio $^{210}Po/^{210}Pb$ seems to increase from March 2003.

1B09: ^{137}Cs in recent fallout samples in relation to the Asian Continent aerosols (2)

Ishikawa, Y., Takahashi, M. (Environ. Radioact. Res. Inst. Miyagi), Narazaki, Y., Tagami, S. (Fukuoka Inst. Health & Environ. Sci.), SUZUKI, T. (Fac. of Sci., Yamagata Univ.)

^{137}Cs deposition (rain and dry fallout) was investigated from July 2002 in Miyagi and Yamagata Prefs., Japan. Relatively high values of ^{137}Cs were observed in Sakata City, Yamagata Pref.. Airborne dust samples collected from mid-March to late-April in 2002 in Miyagi and Fukuoka Prefs., Japan showed high concentrations of ^{137}Cs (about 92 $\mu Bq/m^3$ in max.), during and after the Asian Continental Dust Events (Kosa). This fact directly shows the contribution of the Asian Continent-derived ^{137}Cs to the Japanese atmospheric circumstances.

1B10: Seasonal variation of $^{228}Ra/^{226}Ra$ ratio in seaweed samples of Noto coastal area

Inoue, M., Kofuji*, H., Yamamoto, M. and Komura, K. (K-INET, Kanazawa Univ.)

In order to investigate the distributions and behaviors of radium in the coastal area, sixty-eight seaweed (*Sargasso*) and 27 coastal water samples were collected in Noto Peninsula, Japan during the period from December 1998 to April 2002, and analyzed for the ^{228}Ra and ^{226}Ra activities by low background gamma spectrometry. As results, the ^{228}Ra and ^{226}Ra activities in *Sargasso* samples showed a wide range from 10 to 50 dpm/kg-fresh and 30 to 150 dpm/kg-fresh, respectively. However, it is noteworthy that the $^{228}Ra/^{226}Ra$ activity ratio of *Sargasso* demonstrated the seasonal variation with minimum values ($^{228}Ra/^{226}Ra$ = ~1.2) in June and maximum (2.5 ~ 3.5) in December, which are similar to that of the surrounding seawater. Therefore, we suggest that seaweeds such as *Sargasso* possibly retain the $^{228}Ra/^{226}Ra$ ratio of surrounding seawater and this temporal variation is expected to give knowledges about the seasonal changes of coastal environment.

1B11: Uranium and thorium isotopic composition in lake bottom sediment and environmental changes – Lake Biwa in Japan

Sakaguchi, A.[1], Yamamoto, M.[2], Ishikawa, K.[1], Kashiwaya, K.[2], Ohtsuka, Y.[3] ([1]Graduated School of Natural Sci. and Technol., Kanazawa Univ., [2]K-INET., [3]IES.)

Some sediment cores (0 – ca. 40 cm in depth) were taken from Lake Biwa in Japan, and all samples divided into 1 cm thin fraction were subjected to analyses of U (^{238}U and ^{234}U) and Th (^{232}Th, ^{230}Th and ^{228}Th) isotopes. Lead-210 and ^{137}Cs for estimating the sedimentation rates were also determined along with physical and chemical parameters such as grain size, magnetic susceptibility, water content, biogenic-SiO_2, etc. By using model equations, U found in each fraction of the core were attempted to separate mainly into two different U components due to lithogenous and autogenous fractions. The depth profiles of U and Th isotopes thus obtained were compared each other with those of physical and chemical parameters. In some parts of depth in each core, changes in lithogenous $^{238}U/^{232}Th$ ratios are found to coincide with those of grain size, which is closely related to the change in erodibility of surface soil or rock in the catchment area and rainfall intensity. These results suggest that changes in the lithogenous U and Th isotopic ratios separated by model may be helpful in tracing the past environmental changes surrounding lake area.

2A01: Constructions and physical properties of functional spin transition iron(II) compounds

Hayami, S.,[a] Danjobara, K.,[a] Miyazaki, S.,[a] Shigeyoshi, Y.,[a] Ogawa, Y.,[b] Matsumoto, N.,[b] Inoue, K.,[c] Maeda, Y.[a] ([a]Kyushu Univ., [b]Kumamoto Univ., [c]IMS)

A number of spin-crossover iron (II) compounds have been studied. They are important in the development of electronic devices such as molecular switches. Here, we synthesized the spin-crossover iron(II) compounds $[Fe(R)_2(NCS)_2]$ (**1**) and $[Fe(3C16)_2(NCS)_2]$ (**2**). The iron(II) compounds **1** has exhibited photo-induced spin transition (LIESST) effect. The ligand R in compound **1** has an asymmetric carbon atom, and the compound **1** is a chiral molecule (R form). Generally, chiral compounds exhibit the second-order non-linear optical (SONLO) response. The NLO response is also dependent on the electronic configuration of the metal center. The LIESST iron(II) compounds exhibited spin transition between a paired and an unpaired electronic configurations by illumination can switch the SONLO response. The compound **2** has also exhibited photo-induced spin transition behavior. The ligand 3C16 with elongated substituents were employed to provide the rodlike geometry and the liquid crystal properties of the compound **2**. The compound **2** exhibits liquid crystal phase in the range 345-400 K. This compound is a first example showing photo-induced spin transition and liquid-crystalline properties in the compound.

2A02: Photo magnetic properties of some iron(III) LIESST compounds

Kawahara, T., Hayami, S., Inoue†, K., Maeda, Y. (Kyushu Univ., †IMS)

Several iron(III) LIESST compounds has been reported, until now, although LIESST phenomenon is intensively reported for iron(II) compounds. Here, the iron(III) LIESST compounds $[Fe(R\text{-}pap)_2]X$ were prepared (R = CH_3, Cl, and X = ClO_4, BF_4, PF_6), and counter anion effect and the substituent group effect were investigated in order to make clear the mechanism of LIESST phenomenon for iron(III) complexes. Generally the correlation of the spin transition temperature ($T_{1/2}$) and the relaxation temperature after illumination T_c(LIESST) is inverse, the position of each compounds in the T_c(LIESST)- $T_{1/2}$ diagram has been analysed and discussed. The relation between T_c(LIESST) and $T_{1/2}$ is T_c(LIESST) = $T_0 - 0.6T_{1/2}$.(in this case, T_0 = 226.13), where the value of T_0 is a value of T_c (LIESST) in case $T_{1/2}$ is 0. It is considered that the values of T_0 closely relate to the intermolecular cooperativity.

2A03: Mössbauer spectroscopic study of assembled complexes

Nakashima, S.,[1] Asada, Y.,[2] Yamamoto, A.,[2] Okuda, T.[2] ([1]N-BARD, Hiroshima Univ., [2]Graduate School of Sci., Hiroshima Univ.)

^{57}Fe Mössbauer spectroscopy was applied to the study of the assembled iron complexes and the thermal decomposition phenomena. The Mössbauer parameters revealed a high-spin Fe^{II} state. $\{[Fe(NCS)_2(H_2O)_2(bpy)](bpy)\}_n$ has a large QS value because of FeN_4O_2 arrangement.The QS value decreased by removing water molecule, suggesting FeN_6 arrangement. The value increased again by removing one bpy, suggesting FeN_4S_2 arrangement by bridging with NCS^-.The results support the proposed thermal decomposition.We also synthesized the similar complexes by changing the bridging and NCS^- ligands, which were compared with $\{[Fe(NCS)_2(H_2O)_2(bpy)](bpy)\}_n$.
Fe-*trans*-1,2-bis(4-pyridyl)ethylene-NCS complex showed two new crystal forms. The two forms were also characterized by ^{57}Fe Mössbauer spectroscopy.

2A04: Variable temperature Mössbauer spectroscopic studies on mixed-valence iron dicarboxylic acid complexes

Afroj, D., Katada, M. (Graduate School of Science, Tokyo Metropolitan University)

In a continuation of our study to prepare oxo-centered trinuclear mixed-valence iron dicarboxylic acid complexes two new mixed-valence complexes iron glutarate and iron succinate have been prepared and studied by variable temperature Mössbauer spectroscopy. In this study the complexes showed a trapped to detrapped valence state of high spin Fe(III) and high spin Fe(II) from liquid nitrogen temperature to room temperature. Two distinct doublets correspond to high spin Fe(III) and Fe(II) were observed specifying the slower electron transfer rate then the Mössbauer time scale (10^{-7} s) at liquid N_2 temperature for both the complexes. Spectral line shapes were changed with increasing temperature indicating the existence of relaxation effect associated with the increased intramolecular electron transfer rate in the complexes. At room temperature iron glutarate complex still showed a relaxation spectrum. Iron succinate complex showed an averaged doublet at room temperature with small Fe(III) impurity observed as a shoulder in the spectrum. TG-DTA measurements suggested the presence of solvated water molecules in the complexes.

2A05: Spin-crossover behavior of 2-D iron complexes with imidazole-imidazolate bridges

Iijima, S.,[a] Mizutani, S.,[a] Ikuta, Y.,[b] Oidemizu, M.,[b] Matsumoto, N.,[b] Sunatsuki, Y.,[c] Ohta, H.,[c] Kojima, M.[c] ([a]Natl. Inst. Ind. Sci. Technol., [b]Facl. Sci., Kumamoto Univ., [c]Facl. Sci., Okayama Univ.)

Two types of 2-D iron complexes with imidazole-imidazolate bridges, $[Fe^{II}H_3L^{Me}][Fe^{II}L^{Me}]X$ and $[Fe^{II}H_3L^{Me}][Fe^{III}L^{Me}]X_2$ were investigated by ^{57}Fe Mössbauer spectroscopy, where H_3L^{Me}= tris-[2-(((2-methylimidazoyl-4-yl)methylidene)amino)ethyl]amine. $[Fe^{II}H_3L^{Me}][Fe^{II}L^{Me}]X$ behaved a two-step spin transition indicating different spin-transition temperatures of the two Fe^{II} sites in these complexes. $[Fe^{II}H_3L^{Me}][Fe^{III}L^{Me}]X_2$ showed a spin-transition of the Fe^{II} site on elevating the temperature from 4.2 K, followed by the spin-transition of Fe^{III} site. The temperature dependence of the Fe^{III} site's line shape was based on a "rapid exchange process" between the low-spin and high-spin states of Fe^{III}.

2B01: Background components of Ge detectors in Ogoya Underground Laboratory

Hamajima, Y., and Komura, K. (LLRL, Kanazawa University)

From 1995, ultra low background (BG) counting system has been set up at Ogoya underground laboratory, OUL (270m water equivalent) in the old mining tunnel. There are 5 well, 4 planar, and a coaxial type Ge detectors with low-level background count-rate, with high detector efficiency, and with pure massive shield. Here, we propose new BG value normalized in Ge surface area (cpd/cm^{2}Ge) to compare BG level between different types of Ge detectors. In this BG value, best one was 5.24 for planer type detector (38cm^2 x 3cm) and 5.41 for coaxial detector (93% efficiency) in OUL. Anticoincidence system with plastic scintillator (PS) was tested to reduce cosmic-ray (CR) induced components. Energy spectra of PS with 5cm thickness were measured at 0 and 270 mwe. Coincidence count rate of Ge-PS did not depend on thickness of PS and time. Angular distribution of CR was cosθ or cos^{2}θ as a function of altitude angle of θ. At OUL, BG count rate of Ge-PS coincidence events was 0.4cpd/cm^{2}Ge for both planer and coaxial Ge with effective PS solid angle of 0.2 sr.

2B02: High resolution simultaneous measurements of airborne radionuclides

Komura, K., Muguntha Manikandan M., Yamaguchi, Y. (LLRL, K-INET., Kanazawa Univ.) Iida, T., Moriizumi, J. (Grad. School of Eng., Nagoya Univ.) and Members of 21 Century COE Group

"High resolution simultaneous measurements of airborne radionuclides" was conducted to investigate horizontal and vertical movement of air-mass and aeolian dusts. Rn-222 was measured by high sensitive radon monitors with one hour of intervals. Other airborne radionuclides were collected on filter paper made of quartz fiber at flow rate of 700L/min by high volume air sampler at Tatsunokuchi (Low Level Radioactivity Laboratory) and Hegra Island located 50 km from Wajima in Noto Peninsula. The Hegra Island was chosen as sampling site because area is extremely small (1.2 km^2) and there is no car except few ones for business use. Sampling intervals are 1 to 7 days in usual case, and 1-2 hours at the cases of typhoon and cold (warm) front passing. Concentrations of Pb-210 and Be-7 were measured by ultra low-background Ge detectors and Po-210 was determined by alpha spectrometry after electrodeposition of polonium on silver plate. Meteorological data (wind speed and direction etc.) was obtained from Ishikawa Pref. Inst. of Public Health and Noto Branch of Japan Coast Guard. Outline of the project and preliminary results will be presented together with future plan.

2B03: Radionuclide concentrations in marine sediments off Rokkasho

Kofuji, H., Gasa, S., Kamamoto, M., Mori, M., Nakayama, T., Nishizawa, K., Kuji, T., Iseda, K., Shima, S., Kawamura, H. (Japan Marine Science Foundation)

Concentrations and inventories of radionuclides ($^{239+240}$Pu, ^{90}Sr, ^{137}Cs and $^{210}Pb_{excess}$) in marine sediment samples collected from off Rokkasho were measured. Inventories of $^{239+240}$Pu and $^{210}Pb_{excess}$ in sediments were from 29 to 219 Bq/m2 and 10 to 65 kBq/m2 respectively. Inventories of these nuclides on 2 stations near shore area were about 2 times higher than ones on land near the sea area. It is suggested that these nuclides was supplied from land and/or another station of sea area. Except these 2 stations, it was shown the tendency that inventories were high on the north side and low on the south side in the sea area.

2B04: Present status of development in analytical techniques for safeguards environmental samples

Usuda, S., Watanabe, K., Sakurai, S., Magara, M., Esaka, F., Kohno, N., Takahashi, M., Saito, Y., Esaka, T.K., Miyamoto, Y., Inagawa, J., Yasuda, K., Lee, C.G., Gunji, H., Kurosawa, S., Iguchi, K., Onodera, T., Fukuyama, H., Ichimura, S., Suzuki, D., Imai, J., Chai, J.Y., Doan, T.M. (JAERI)

JAERI is promoting a project to establish the environmental sample analysis techniques for the IAEA strengthened safeguards system. The construction of a clean room laboratory (Clean Laboratory for Environmental Analysis and Research: CLEAR) was completed in June 2001. At CLEAR, essential techniques for the environmental bulk and particle swipe sample analysis were established as ultra-trace analytical method of uranium and plutonium. In addition, similar bulk analysis techniques for hot-cell swipe samples are also being developed at NUCEF. In January 2003,

JAERI was qualified as a member of IAEA environmental network analytical laboratories. Hence the second phase of the project has just started in order to develop more advanced techniques, which aim at analyzing minor actinides and fission products as well as uranium and plutonium. In near future, analysis of domestic and IAEA samples will be started after contracting with IAEA. This paper deals with recent topics on the developed techniques for bulk, particle and hot-cell sample analyses, and future perspectives.

2B05: Application of FT-TIMS method to environmental sample analysis for safeguards

Esaka, T.K., Esaka, F., Inagawa, J., Onodera, T., Iguchi, K., Lee, C.G., Sakurai, S., Usuda, S., Watanabe, K. (JAERI)

Secondary ion mass spectrometry (SIMS) is commonly used for the isotope ratio analysis of individual uranium particles in safeguards environmental samples, however, the SIMS analysis of the particles with diameters less than 1 μm is difficult due to its insufficient sensitivity. The authors investigated the combination of fission track (FT) method and thermal ionization mass spectrometry (TIMS) to realize the sensitive detection and isotope ratio analysis of such uranium particles. The swipe samples used for this study were prepared from the particles of NBS U350 standard reference materials. The particles on the sample were recovered and entrapped into a track detector film made of polycarbonate. The film was irradiated with thermal neutrons in the JRR-4 reactor at JAERI, where tracks were created by U-235 fission fragments. After cooling, the film was etched with 6M NaOH for 8 minutes at 55℃. The uranium particles were successfully found by the observation of the tracks, and the uranium isotope ratios of the individual particles were measured with TIMS. These results suggest the feasibility of the FT-TIMS method.

3A01: Synthesis and ^{237}Np Mössbauer Spectra of Dioxobis(2,4-pentanedionato)(pyridine) neptunium(VI)

KawasakI, T.,[*,‡] Nakada, M.,[*] Saeki, M.,[*] Kitazawa, T.,[‡] Takeda, M.[‡] ([*] Jpn.At.En.Res. Inst., [‡]Fac. Sci. Toho Univ.)

The complex was synthesized from acetonitrile solution of neptunyl(VI) nitrate by adding excess pyridine and acetylacetone, directly, in glove box at room temperature. The characterization was made by XRD. ^{237}Np Mössbauer spectra were measured at 11, 20, 30 and 40 K. The 16 absorption lines due to quadruple and magnetic splitting were observed.

3A02: Correlation between thermal annealing behavior of radiation defects and tritium release behavior in neutrton-irradiated solid tritium breeding materials

Oyaidzu, M.,[a] Kimura, H.,[a] Kawamoto, K.,[b] Munakata, K.,[c] Okada, M.,[b] Nishikawa, M.,[c] Okuno, K.[a] ([a]Radiochemical Res. Lab., Fac. of Sci., Shizuoka univ., [b]Res. Reactor Inst., Kyoto univ., [c]Dept of Adv. Energy Eng. Sci., Kyushu univ.)

In D-T fusion reactors blanket system, hot tritium would be produced mainly by the reaction ^{6}Li(n,□)T. Therefore, it is very important to interpret the tritium existence states and behavior to establish the tritium recovering system. Li_2TiO_3, the sample of this study, is one of candidates for solid tritium breeding materials. Annihilation processes of radiation defects induced by neutron irradiation were observed by means of ESR (Electron Spin Resonance), because tritium release processes seem to be correlated with that, from the view of previous study of other solid tritium breeding materials. Annealing experiments indicate that there are two annihilation processes of radiation defects. The two processes, the fast process and the slow process, seem to have concern with lithium and oxygen diffusion, respectively. When the annihilation behavior of radiation defect is compared with tritium release behavior, the slow process, oxygen diffusion, has concern with tritium release.

3A03: Coordination geometry of tricarbonyl technetium(I) complexes with salen type ligands

Harano, A., Takayama, T., Sekine, T., Kudo, H. (Tohoku Univ.)

Novel tricarbonyl technetium(I) complexes with salen type Schiff base ligands (H_2salen and H_2dmsalen) and aminephenol ligands (H_2hben and H_2dmhben) were synthesized from ligand substitution reactions of $(Et_4N)_2[Tc^{I}Cl_3(CO)_3]$. These complexes were characterized by ^{1}H NMR and IR spectroscopies. In ^{1}H NMR spectrum, signals for ethylene and benzene groups of a ligand in the salen complex showed a downfield shift as well as a change of spin multiplicity. This complex showed C=O and C=N stretching bands in the IR spectrum, indicating the formation of $[Tc^{I}(CO)_3(Hsalen)]$. In this complex, the salen ligand coordinates to the Tc atom with two N atoms and one O atom. For H_2dmsalen, a technetium complex was not formed due probably to a steric hindrance of methyl groups in coordination to technetium. On the other hand, H_2hben and H_2dmhben formed $[Tc^{I}(CO)_3(Hhben)]$ and $[Tc^{I}(CO)_3(Hdmhben)]$. For H_2dmhben, the rotation of the C-N single bond can moderate the steric hindrance of methyl groups, leading to the formation of the complex.

3A04: Structure and dπ-orbital energy of $Tc^{IV}Cl_2$(salen) type complexes

OMICHI, K., KODO, R., TAKAYAMA, T., SEKINE, T., KUDO, H. (Tohoku Univ.)

Structures and redox properties of technetium(IV) complexes with salen type Schiff base ligands such as salen, salpn, salbn, dcsalen, dmsalen and dbsalen were examined. X-ray crystallography for these complexes showed that these complexes were in an octahedral structure with two *trans* chloro ligands and a tetradentate salen ligand. The magnetic moment of these complexes measured by the Evans method indicated that the technetium was in the +4 oxidation state with three unpaired electrons. Cyclic voltammograms of these complexes showed a reversible Tc(IV)/Tc(III) redox wave. The redox potential was lower in the complex with electron donating substituents or long alkyl chains in the ligand, compared with that in the other complexes examined here. The result indicates that the dπ-orbital of the technetium complexes becomes higher in energy when electron donating substituents are introduced in the ligand or the alkyl chain of the ligand is elongated.

3A05: A factor governing the formation ratio of *syn* to *anti* isomers of technetium(V) complexes of PnAO ligands with benzyl group in bridging alkyl chain

Takayama, T., Morimoto, Y., Sekine, T., Kudo, H. (Tohoku Univ.)

Formation ratios of *syn* to *anti* isomers of nitridotechnetium(V) complexes as well as the oxotechnetium(V) complexes of PnAO (3,3,9,9-tetramethyl-4,8-diazaundecane-2,10-dione dioxime) derivatives with a benzyl group were examined. The formation ratios between *syn* and *anti* isomers of the nitridotechnetium(V) complexes with substituted PnAO were almost 50 to 50, while the oxotechnetium(V) complexes preferred the *anti* isomer. A stereoselectivity for the formation of technetium complexes with substituted PnAO depends on the steric repulsion between the substituent and the terminal ligand. The formation ratio of *syn* to *anti* isomers of the nitrido complexes without steric repulsion is unity, but the steric repulsion in the oxo complexes makes it preferable to form the *anti* isomer. The difference in the steric effect between the nitridotechnetium(V) and oxotechnetium(V) complexes arises from the conformation of the six-membered chelate ring that includes a propylene bridge.

3A06: Evolution of the multitracer production technology on the RI beam factory project

Enomoto, S., Haba, H., Hirunuma, R., Igarashi, K., Yano, Y. (RIKEN)

In 1991, we have established a multitracer technology, which enables the simultaneous tracing of a number of elements in various biological and chemical systems. The e RIKEN multitracer technology has been successfully applied to investigators of behavior of various elements in the field of bio-trace elements research. Recently, we developed the new RI production system such as a gas-jet-coupled multitarget system, the single radioisotope production system by using the AVF cyclotron and the RI cocktail production unit on RIKEN Projectile Fragments Separator. In this paper, we describe the usefulness of the multitracer technology on next generation for studies on trace elements in animals and plants.

3A07: Change of concentrations of trace elements in pancreatic cell of zinc deficiency mice

Kinugawa, N., Ohyama, T., Ogi, T., Kajita, Y., Noguchi, M., Suganuma, H., Yanaga, M. (Fac. of Sci., Shizuoka Univ.)

The distribution of various trace elements in to pancreatic cell of Zn-deficient mice and control mice was investigated. Eight-week old male mice of ICR strain were divided into two groups; one was fed with Zn-deficient diets and the other with control diets, respectively. After three weeks, their pancreata were removed and homogenized with HEPES buffer. The homogenates were separated into nucleus, mitochondria, microsome, and cytosol by differential centrifugation at 1,000×g for 20 min, 9,000×g for 20 min and 105,000×g for 65 min, successively. Concentrations of 10 elements, Na, Mg, Cl, Mn Fe, Co, Zn, Se, Br and Rb, in each subcellular fraction were determined by an instrumental neutron activation analysis. The zinc concentrations in all subcellular fractions of Zn-deficient mice were lower than those of the control mice. On the other hand, the Co concentrations in nucleus and cytosol fractions of Zn-deficient mice were higher than those of the control mice.

3A08: Searching for the metal binding factor responsible for involved in a variation of concentration of trace elements under the Zn-deficient condition

Ohyama, T.,[a,b] Watanabe, K.,[c] Kinugawa, N.,[a] Yanaga, M.,[a] Noguchi, M.,[a] Suganuma, H.,[a] Ishikawa, K.,[a] Hirunuma, R.,[b] Takahashi, K.,[b] Enomoto, S.[b] ([a]Faculty of Science, Shizuoka University, [b]RIKEN, [c]NIRS)

In this work, we attempted to analyze the affinity between the trace elements and Zn binding proteins which induce the variation of the concentrations of trace elements under the Zn-deficient condition. Eight-week old male mice were divided into Zn-deficient and control groups. After three weeks treatment, the supernatant fractions of their livers were divided into 35 fractions by gel filtration chromatography. The concentrations of trace elements and the protein amounts in each fraction were determined by ICP-MS and BCA protein assay method, respectively. The solutions of fractions which have high values of Zn concentration / protein amount ratio were poured into centrifugal filter tubes, and then, a multitracer solution was added. After the centrifugation, the radioactivities in filters and filtrates were measured by HP Ge detectors. The radioactivities of Mn, Zn and Co in filter of 15th fraction of Zn-deficient mice were higher than those of control ones. This indicates the existence of proteins which have a great affinity to these elements in this fraction.

3B01: Changes of luminescence property from quartz accompanied with thermal treatment in oxidative condition

Yamaguchi, T.,[a] Tazika, Y.,[b] Hashimoto, T.[b] ([a]Graduate School of Sci. and Technol., [b]Niigata univ., Fac. of Sci., Niigata univ.[1])

Z-cut slice samples of original and annealed quartz, prepared from Madagascan crystal rock, were observed with color center image (CCI) and thermoluminescence color image (TLCI) after γ-ray irradiation. It was found from original quartz slice that intense and weak BTL patterns on the TLCI correspond to the dense and light gray color on the CCI, whereas TL signals from annealed quartz enhanced greatly their sensitivities in comparison with the original quartz. Concerning the luminescence sensitizations, the distribution patterns due to OH-impurities from the IR absorption spectra gave almost opposite tendency to the color center images (CCI) and the TLCI on original slice. After annealing treatments, some Li-dependent OH absorption completely disappeared by accompanied with enhancing BTL-sensitivities. From results of IR absorption spectra, ESR spectra and low temperature TL glowcurves, OH impurities, including Al-OH, Li dependent OH and H_2O, decreased after thermal treatment. Enhancement of such luminescence signal must be responsible for decrease of H^0 radical, formed as a radiolysis product of OH impurities.

3B02: Influence of some kinds of radiation on luminescence property from white minerals

Yawata, T. (Graduate School of Science and Technology, Niigata University), Shimizu, N., Hashimoto, T. (Department of Chemistry, Niigata University)

Radiation induced luminescence phenomena, such as thermoluminescence (TL), radioluminescence (RL) and so on, have been observed, when quartz grains are irradiated with ionizing radiations. Nowadays, the usefulness of TL-dating techniques to burnt archaeological materials is widely recognized. Especially red thermoluminescence (RTL) of natural quartz grains is known to offer preferable properties for Quaternary dating.An artificial and standard radiation source, which serves as a dose-normalization to natural radiations, is necessary for the determination of accumulated doses using luminescence measurements. Authors have so far proposed the use of X-ray generator as a standard source instead of RI source. The X-ray generator exhibits several excellent properties, such as (i) good linear relationship between tube current and dose rate, (ii) wide dynamic range of applicable doses, and (iii) highly uniform irradiation to sample area. In this study, the optimal irradiated conditions were searched by X-ray spectrometry with germanium semiconductor detector, changes of aluminum-absorber thickness to exclude low-energy components, and RTL measurements using each quartz single grain irradiated by this X-ray generator. Consequently, it was found to attain the preferable dosimetry by means of the aluminum-absorber over 50 μm thickness.

3B03: Property changes of red thermoluminescence (RTL) and red infrared-stimulated luminescence (R-IRSL) from feldspars annealed in either oxidative or reductive condition

Mitamura, N.,[a] Hashimoto, T.[b] ([a]Graduate School of Science and Technology, Niigata University., [b]Faculty of Science, Niigata University)

The feldspars are expected to be hopeful for luminescence dating because most feldspars display a luminescence signal brighter than that from quartz. When infrared light was illuminated on feldspar slices after X-ray irradiation, infrared stimulated luminescence color images (abbreviated to IRSL-CI) showed three color patterns, separable into intense greenish yellow, red and faint violet portions. The property changes of R-IRSL and RTL in two kinds of feldspar (albite and microcline) grains were investigated by applying thermal annealing treatments in oxidative or reductive condition for 10h at 600°C, 800°C and 1000°C. In addition to each luminescence measurement, ESR signals related to Fe (III) were measured. The annealed sample in oxidative condition above 800°C showed obvious enhancement of R-IRSL and RTL sensitivities, coincident with the increasing ESR signals due to Fe (III), in comparison with keeping the almost stationary luminescence

intensity from original or reduced samples. As a result, the enhancement in the red emission regions by oxidative annealing supports some contributions of Fe (III) in feldspar.

3B04: Development of new infrared stimulated luminescence in red-regions (R-IRSL) from feldspar for dating of archaeologically burnt materials

Usuda, H., (Graduate School of Science and Technology, Niigata Univ.), Nakata, Y., Hashimoto, T., (Faculty of Science, Niigata Univ.)

Radiation induced luminescence phenomena, such as thermoluminescence (TL) and optically stimulated luminescence (OSL), are known to be effective in widespread use for archaeological and quaternary geological dating as well as for a retrospective dosimetry using sediment layers and burnt materials. Among these luminescence measurements, the luminescence sensitivity of feldspar is overwhelmingly intense in comparison with that of quartz, although there is anomalous fading effect in feldspar. In this situation, a new detection aimed at far-red (700~800nm) emission (infrared stimulated luminescence in red-regions, abbreviated to R-IRSL) has been developed because of assuming negligible contribution of anomalous fading. For the detection of this emission, firstly the best condition of R-IRSL measurements was searched for the sake of reducing the background as low as possible by making choice of stimulating IR-LED, PMT, and combination of optical filters in this IRSL-emission system. On the basis of the best condition of R-IRSL, the probability of R-IRSL dating was confirmed from dose response curves.

3B05: Radioluminescence properties from white minerals

Fujita, H.,[a] Hashimoto, T.[b] ([a]JNC, [b]Niigata Univ.)

When an insulating white mineral was exposed to ionizing radiation, both electrons and positive holes are simultaneously created, fixed portion can be trapped as defects in the crystal lattice. Subsequently, when the thermal or the optical stimulation was applied to high energy levels enough to elevate the electrons to the conduction band, the electrons can recombine with luminescent centers by giving light emission. Thus, thermoluminescence (TL) and optically stimulated luminescence (OSL) phenomena have been utilized to the radiation-dosimetry. However, these luminescence measurements are necessary to a heat and a light sources on the stimulation stage. On the other hand, radioluminescence (RL) phenomena, which can be observed during the irradiation without any stimulation sources, attract much attention to both the alternative dosimetry and the elucidation of luminescence mechanism. RL-detection equipment was developed to follow the RL-property changes of quartz samples during irradiation. The blue-RL (B-RL) intensity tends to a saturating tendency at the exposure time after about 100 seconds, subsequently giving slightly increasing intensities, while red-RL (R-RL) entirely showed linear increasing trend at least up to 500 seconds.

3B06: Determination of ^{36}Cl in soil environment by AMS

Matsuhiro, T.,[a] Seki, R.,[a] Nagashima, Y.,[a] Takahashi, T.,[a] Sasa, K.,[a] Miura, T.,[b] Bessho, K.,[b] Matsumura, H.,[b] Wang, Q.,[b] Ono, S.,[a] Sueki, K.,[a] Endo, Y.[a] ([a]AMS group of the University of Tsukuba, [b]KEK)

Chlorin-36 is a very long half-lived radio-nuclide, $T_{1/2}$ = 3.01×10^5 years, and is produced mainly by spallation of heavier nuclei, principally Ar in the atmosphere by energetic cosmic ray. As the other origin, ^{36}Cl is produced by thermal neutron activation of ^{35}Cl by nuclear tests, and emission of ^{36}Cl produced in nuclear reactor. As the natural existence of ^{36}Cl is a very low, so accelerator mass spectrometry (AMS) is only available method for measuring the nuclide. In the measurement, the ^{36}S isobar causes a significant error. Therefore, the removal of sulfur contaminant in the sample is necessary requested. So, we have developed a very efficient procedure for eliminating the sulfur contaminant from the sample. A critically accident occurred on September 30, 1999 at the facility JCO in Tokai-mura. Rather high ratios of ^{36}Cl/^{35}Cl in soil collected in and around the JCO facility were measured. The highest ratio in Tokai-mura was about hundred times larger than in Tsukuba. For searching the reason, we have measured ratios of ^{36}Cl/^{35}Cl in soil samples collected in various areas in Japan.

3B07: AMS measurement of ^{36}Cl induced in shielding concrete of various accelerator facilities

Bessho, K.,[a] Matsumura, H.,[a] Matsuhiro,[b] T., Miura,[a] T., Wang,[a] Q., Masumoto, K.,[a] Nagashima, Y.,[b] Seki, R.,[b] Takahashi, T.,[b] Sasa, K.,[b] and Sueki, K.[b] ([a]KEK, [b]AMS Group, Univ. of Tsukuba)

The concentrations of ^{36}Cl induced in shielding concrete of the various accelerators has been measured by accelerator mass spectrometry. For four kinds of accelerator facilities, SF cyclotron (CNS, Univ. of Tokyo), 300 MeV electron LINAC (LNS, Tohoku Univ.), 1.3 GeV electron synchrotron (Tanashi Branch, KEK) and 12 GeV proton synchrotron (KEK), the depth profiles of ^{36}Cl / ^{35}Cl ratios in concrete samples near the beam lines were analyzed. The depth profiles of ^{36}Cl / ^{35}Cl are consistent with those of the radioactive concentrations of ^{152}Eu and ^{60}Co, which are formed by thermal neutron capture reactions. These results imply that ^{36}Cl formed in shielding concrete of these accelerators is mainly produced by thermal neutron capture of ^{35}Cl. The maximum ^{36}Cl / ^{35}Cl ratio corresponds to the specific radioactivity of 2×10^{-3} Bq/g, which is not serious for radioactive waste management in reconstruction or decommissioning of accelerator facilities.

3B08: Analysis of tritium in shielding concrete of various accelerator facilities

Wang, Q., Bessho, K., Masumoto, K., Matsumura, H., Miura, T., Shibata, T. (KEK)

Many radioisotopes are induced in the shielding concrete of accelerator facilities by secondary neutrons. Within radioisotopes in activated concrete, the tritium is difficult to determine because of its decay by emitting only low-energy beta-rays. The objective of this study is to establish a rapid, simple and quantitative method for analyzing tritium in shielding concrete samples of accelerator facility. As the usual heating method using an electric furnace is time-consuming, we tried to use an IR-furnace for tritium extraction. After being studied on the collection conditions regarding heating temperature and heating time, tritium in concrete samples for three kinds of accelerator facilities were measured. The thermal neutron flux in an accelerator room during operation was estimated by using tritium activity and Li concentration determined by Atomic Absorption Spectrometry. Obtained neutron fluxes were almost same as that obtained by the relation between ^{60}Co and Co concentration. It was concluded that tritium was induced by the thermal neutron reaction, because the depth profile of tritium in concrete was almost same as that of ^{60}Co, ^{134}Cs and ^{152}Eu.

POSTER PRESENTATIONS

1P01: Basic study of the mass spectrometry using a tunable laser

Ishizu, H., Nagashima, M., 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. However, the chemical species involved in gas phase chemistry are ambiguous even for their homologs. A mass spectrometry using a tunable laser is examined for the identification of chemical species. In this work, as a basic study for laser ionization, we fabricated an ion detector and examined its performance.

1P02: Development of apparatus for preparing α

source in on-line chemical separation

Saito, J., Hirai, T., Goto, S., Kudo, H. (Fac. of Sci., Niigata Univ.)

In order to prepare thin and uniform α sources for an aqueous chemistry of transactinide elements, we constructed an apparatus for this purpose and examined the possibility of a sample preparation technique by spraying a liquid sample in vacuum. The collection efficiency under various conditions will be presented.

1P03: Development of an automated on-line chemical separation system for single atom chemistry

Hasegawa, H.,[1] Tani, Y.,[1] Toyoshima, A.,[1] Shigekawa, M.,[1] Takahashi, N.,[1] Sato, W.,[1] Yuki, H.[2] Ohtsuki, T.,[2] Takamiya, K.,[3] Shibata, S.,[3] Shinohara, A.[1] ([1]Grad. School of Sci., Osaka Univ., [2]Grad. School of Sci., Tohoku Univ., [3]Res. Reactor Inst., Kyoto Univ.)

We are interested in the chemical behavior of short-lived heavy and trans actinides, which are available in quantities of only a few atoms at a time throughout the accelerator on-line experiments. Our group has designed such systems for the study and tested repeatedly using the spontaneous fission products of ^{252}Cf. In the present study, we have renewed the system to make it applicable to more flexible chemical treatments. We have also introduced a highly sensitive RI-detection system with an alpha-detecting liquid scintillation counter. Results obtained using the system are to be compared with the behavior of elements in macroscopic quantities so as to provide insight into the dependence of the chemical behavior on the number of atoms.

1P04: Study of behaviors of zirconium tetrachloride and oxychloride in gas phase using mass spectrometer

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

The first trans-actinide element Rf (Z = 104) have investigated by rapid gas phase separations, because its halides are volatile like its homologues Zr and Hf. However, the volatility behaviors of their halides in gas phase are still ambiguous, especially under the conditions of oxygen contaminant. Therefore, in this work, $ZrCl_4$ and $ZrOCl_2$ as Zr compound were examined in gas phase using mass spectrometer (MS). Vaporized samples were transported to MS using He gas and vapor pressures were measured as a function of temperature.

1P05: Problems of a chlorinating reagent for gas-phase chemistry experiments of Rf chloride

Goto, S.[a], Kaneko, T.[b], Kudo, H.[a] ([a]Niigata Univ., [b]JAERI)

Carbon tetrachloride, CCl_4, has been used as a chlorinating reagent in a gas-phase chemistry experiment for Rf chloride, since it is also used industrially and scavenges oxygen which produces a nonvolatile oxide and oxide-chloride. In spite of high chlorinating efficiency, it is problem that decomposed products of CCl_4 are produced and cristalize at low temperature. In the present work, we identified these products by a mass spectrometer and discussed the influence on the gas-phase chemistry. The decomposed products were produced appreciably at 700–1000 °C and deposited at a water-cooled trap. The main component is hexachlorobenzene, C_6Cl_6 (melting point 230 °C, boiling point 322 °C). Because the temperature of gas-chromatograph column is perfomed under 400 °C in the actual chemistry experiment, C_6Cl_6 may form an aerosol and transport nuclei of interst.

1P06: Fission Modes Calculated with 3-Dimensional Langevin Equation

Ichikawa, T.,[a] Asano, T.,[b] Wada, T.,[b] Ohta, M.[b] ([a]JAERI, [b]Konan Univ.)

We investigate fission modes of ^{270}Sg with 3-dimensional Langevin equation and discuss the contribution of the dynamical effect to the fission mode. The static analysis for the fission mode by using the potential energy surface is not sufficient to understand the experimental mass-energy distribution. The dynamical calculation taking account of the shell effect is needed in order to understand fission paths. We solve the 3-dimensinal Langevin equation numerically in the potential energy surface including the shell correction and compare the mass-energy distribution with the experiment. From the analysis of our results, we expect the existence of several modes in the fission fragments of ^{270}Sg. We show the dynamical effect plays an important role with respect to the components of the asymmetric mode with low TKE.

1P07: Search for synthesis condition of Hf-fullerenes

Sueki, K.,[a] Akiyama, K.,[b] Kikuchi, K.,[c] Katada, M.[c] ([a]Department of Chem., Univ. of Tsukuba, [b]JAERI, [c]Graduate school of Science, Tokyo Metro. Univ.)

The aim of work was searched for the most suitable synthesis condition of Hf-fullerenes. It was examined that the metallofullerenes synthesis by dc-arc method was pressure dependence under the He atmosphere. And, it was reported that a higher yield of metallofullerenes were performed by carbon rod including purpose metal adding Ni metal. It was confirmed that the yields of Hf-fullerenes took an influence due to the existence of Ni metal. The determination of Hf content of each material (for example soot, crude and HPLC fraction etc.) was performed by neutron activation analysis. It was success to find the condition which get yields of 2-3 times until now.

1P08: Development of production methods of endohedral ^{133}Xe-fullerol

Watanabe, S.,[a] Ishioka, N. S.,[a] Kudo, H.,[b] Shimomura, H.,[c] Muramatsu, H.,[d] Kume, T.[a] ([a]JAERI, [b]Tohoku Univ., [c]Faculty of economics, Shinshu Univ., [d]Faculty of Education, Shinshu Univ.)

Endohedral ^{133}Xe-fullerol ($^{133}Xe@C_{60}(OH)_x$) was synthesized by addition reaction of hydroxyl groups to endohedral ^{133}Xe-fullerene ($^{133}Xe@C_{60}$) for medical application as the therapy for cancer of a bone. The *o*-dichlorobenzene solution containing $^{133}Xe@C_{60}$ was stirred with tetrabutylammonium hydroxide as catalyst and KOH solution. The product was characterized by IR spectroscopy and HPLC. Since the IR spectrum shows a C-O stretching absorption at about 1050 cm^{-1}, the formation of fullerol ($C_{60}(OH)_x$) was confirmed. From the HPLC analysis of the product, the formation of the $^{133}Xe@C_{60}(OH)_x$ was corroborated, showing a strong correlation between $C_{60}(OH)_x$ and ^{133}Xe. From this fact, it was found that $^{133}Xe@C_{60}(OH)_x$ could be synthesized by the production method described in the present study.

1P09: The new irradiation system for multipurpose use in CYRIC at Tohoku Univ.

Yuki, H.,[a] Ohtsuki, T.,[a] Shinozuka, T.,[b] Fujita, M.,[b] Iwata, R.[b] ([a]LNS, Tohoku Univ., [b]CYRIC, Tohoku Univ.)

An automated target transport system was developed for multipurpose use of the RI -production beam course in Cyclotron and Radioisotope Center (CYRIC) at Tohoku University. A target can automatically be transported between the draft chamber in the hot laboratory and the irradiation site in the end of beam course 1 within 2 minutes. The system is operating effectively in the study for nuclear chemistry, nuclear structure, radiopharmacy, hot atom chemistry, charged particle activation analysis and production of radioactive tracers. A helium-jet transport system was also installed in the same beam course for the study of decay properties in heavier elements.

1P10: Excitation energy dependence of mass and total kinetic energy distributions for the asymmetric fission mode in proton-induced fission of uranium isotopes

Nishinaka, I., Tanikawa, M.[1], Goto, S.[2], Nishio, K., Tsukada, K., Asai, M., Nagame, Y. (JAERI, [1]Univ. Tokyo, [2]Niigata Univ.)

Velocities of fragment pairs in the 10 - 13 MeV proton-induced fission of ^{233}U and ^{238}U were measured by a double time-of-flight detection technique. Primary fragment mass and total kinetic energy were derived from the observed velocities. In the asymmetric fission mode, the yield at A ~ 130 is enriched compared with that at A ~ 140 - 145 as excitation energy increases. No theory is related to observed excitation energy dependence. The fragments with A ~ 130 and A ~ 140 – 145 correspond to the spherical shells of Z = 50 and N = 82 and the deformed shell of N = 86 - 88, respectively. In view of shell structure of fragments as well as fissioning nuclei, we will discuss the mechanism of excitation energy dependence of mass and total kinetic energy distributions in the asymmetric fission mode, comparing data for various fissioning systems.

1P11: Charge distributions of fragments in heavy-ion-induced fission

Nishinaka, I., Tanikawa, M.[1], Sueki, K.[2], Nagame, Y. (JAERI, [1]Univ. Tokyo, [2]Univ. Tsukuba)

Charge distributions of secondary fission fragments in the ^{16}O + ^{209}Bi reaction were deduced from the relations between the independent or cumulative yields determined radiochemically and the mass yields obtained by measuring time-of-flight and kinetic energy of fragments. The most probable atomic number in an isobaric chain was derived from the experimentally determined yields of secondary fragments. Assuming charge distributions of primary fragments to obey the minimum potential energy (MPE) hypothesis or the unchanged charge distribution (UCD) one, the most probable atomic number and the mass number of secondary fragments give the most probable mass number and the average number of emitted neutrons for the corresponding primary fragments. Comparing derived dependence of neutron multiplicity on fragment mass for the MPE and UCD hypotheses with those in other symmetric fission of pre- and light-actinides, charge distributions of primary fragments can be more reasonably explained by the MPE hypothesis rather than the UCD one.

1P12: Measurement of production cross sections for 12 GeV proton reaction with Hg, Pb, Bi targets used as spallation neutron source

Numajiri, M.,Miura, T., Suzuki, T. (Radiation Sci. Center, High Energy Accelerator Res. Organization), Kondo, K. (Applied Res. Lab., High Energy Accelerator Res. Organization)

Irradiation experiments were performed in order to investigate the production cross-sections of residual nuclei by proton induced reactions at KEK proton synchrotron facility. Production cross-sections in Hg, Pb, Bi targets, which will be used as spallation neutron sources, were measured by gamma-ray spectroscopy. The present data are compared with the previous results for other heavy elements.

1P13: Development of a new target system for online producing multitracer

Yatsukawa, M.,[a] Kasamatsu, Y., [a] Kikunaga, H.,[b] Kinoshita, N.,[b] Shibusawa, S.,[c] Hashimoto, T.,[b] Arai, M.,[c] Ninomiya, K., [a] Yokoyama, A.,[c] Sato, W.,[a] Takahashi, N.,[a] Shibata, S.,[d] Shinohara., A.[a] ([a]Grad. School of Sci., Osaka Univ.; [b]Grad. School of Natural Sci. and Tech., Kanazawa Univ.; [c]Faculty of Sci., Kanazawa Univ.; [d]National Inst. of Radiological Sci.)

A multitracer technique enables us to trace simultaneously a number of elements in various biological and chemical systems. A heavy ion reaction in the intermediate energy region is useful for producing multitracer elements. For wider application, we have designed a special irradiation apparatus, by which the recoil products are directly introduced into liquid and then transported in a tube from the irradiation chamber to a liquid reservoir placed in front of a Ge-detector. Irradiation experiments were performed with HIMAC (Heavy Ion Medical Accelerator in Chiba) at NIRS (National Institute of Radiological Sciences). We used Nb-foil as a target, and water, pH5 and pH3 acetic acid were tested as catcher solvent. After irradiation, some radioisotopes were detected in the catchers. We compared their yields of recoil products from the targets.

1P14: Liquid-liquid microbatch extraction system for short-lived radionuclide chemistry

Sasaki, T.,[a] Takamiya, K.,[b] Shibata, S.,[b] Moriyama, H.[a] ([a]Dept. Nucl. Technol., Kyoto Univ., [b]Res. React. Inst., Kyoto Univ.)

The liquid-liquid microbatch extraction system was developed for the chemical research of short-lived radionuclides, such as heavy actinides. The equipment is composed of three simple extraction cells for phase mixing, phase separation, and solvent collection with the usage of several dozen microliters. Extraction could be completed in a single operation lasting only a minute, from solvent extraction to preparation for alpha- or gamma-ray spectrometry. In order to evaluate the applicability of the system, the extraction of uranium by tributylphosphate (TBP) was investigated. An aqueous phase of 20 μL containing 10^{-3} mol dm^{-3} UO_2^{2+} in 4 M HNO_3 was shaken vigorously with 20 to 80 μL of dodecane-30% TBP pre-saturated with HNO_3 in the mixing cell. While the apparent extractability of uranium achieved over 97% independent of the volume of organic phase, the total recovery decreased when the volume of organic phase decreased. Each volume of the loss of organic phase immersed by the membrane and the handling loss, e.g. volatilization, was estimated to be 6.3 ± 0.9 and 0.8 ± 1.4 μL, respectively.

1P15: Development of gas jet transport system using dry ice powder

Takamiya, K.,[1] Manabe, K.,[2] Shibata, S.[1] ([1]Res. Reactor Inst., Kyoto Univ., [2]Faculty of Engineering, Kyoto Univ.)

A gas jet transport system for recoiled atoms using dry ice powder was developed. The transporting efficiency of this system was measured using spontaneous fission products of ^{252}Cf (200 kBq). The CO_2 siphon cylinder, the F.P. chamber (^{252}Cf source inside), and the collection chamber were connected by plastic tubes. Dry ice powder flowed with CO_2 gas caught fission products and was transported to the collection chamber. The transporting efficiency was 41%. The experiment with non-siphon cylinder instead of CO_2 siphon cylinder was also carried out in order to confirm the effect of dry ice powder. The efficiency of this system was 7%, and the effect of dry ice powder was confirmed.

1P16: Observation of the disintegration of the ultra low-lying isomer ^{229m}Th

Kasamatsu, Y.,[1] Kikunaga, H.,[2] Takamiya, K.,[3] Mitsugashira, T.,[4] Nakanishi, T.,[5] Ohtsuki, T.,[6] Yuki, H.,[6] Sato, W.,[1] Takahashi, N.,[1] Yokoyama, A.,[5] Yamana, H.,[3] Ohkubo, Y.,[3] Hara, M.,[4] Kimura, H.,[1] Iwasaki, M.,[1] Ninomiya, K.,[1] Yatsukawa, M.,[1] Tani, Y.,[1] Hasegawa, H.,[1] Shibata, S.,[3] Kawase, Y.,[3] Shinohara, A.[1] ([1]Grad. School Sci., Osaka Univ., [2]Grad. School Nat. Sci. Tech., Kanazawa Univ., [3]Res. Reactor Inst., Kyoto Univ., [4]The Oarai-branch, Inst. Materials Res., Tohoku Univ., [5]Faculty Sci., Kanazawa Univ., [6]Lab. Nucl. Sci., Grad. School Sci., Tohoku Univ.)

A Low-lying isomer ^{229m}Th is expected to have a unique property that the decay process varies depending on its chemical state; it could be a good subject of the investigation of an electron bridge mechanism.We have tried to produce ^{229m}Th by various nuclear reactions such as $^{228}Ra(n,\gamma)^{229}Ra$, $^{230}Th(\gamma,n)^{229}Th$, $^{232}Th(\gamma,p2n)^{229}Ac$, $^{230}Th(p,d)^{229}Th$ and $^{232}Th(p,p3n)^{229}Th$. Photon measurements and α-particle measurements have been performed to investigate the decay property of ^{229m}Th. In the present work, we report the results of the measurements with a newly developed photon measurement system as well as those of the α-particle measurements for ^{229m}Th.

1P17: Measurements of electronic X rays correlated

with pionic X rays

Ninomiya, K., Kasamatsu, Y., Goto, K., Kikunaga, H., Shigekawa, M., Kinoshita, N., Tani, Y., Hasegawa, H., Yatsukawa, M., Sato, W., Takamiya, K., Matsumura, H., Yokoyama, A., Sueki, K., Hamajima, Y., Miura, T., Shinohara, A. (Grad. School Sci., Osaka Univ., Grad. School Natural Sci., Kanazawa Univ., Res. Reactor Inst., Kyoto Univ., Res. Center Radiation, KEK, Faculty Sci., Kanazawa Univ., Dept. Chem., Univ. Tsukuba, LLRL, Inst. Nature and Env. Technol., Kanazawa Univ.)

In the previous studies, the LMM model of the pion capture process was developed by the experimental facts based on the measurements of pionic X rays and neutral pion decays. We are aiming at investigating the microscopic and dynamic process of the pion capture by examining the state of atoms or molecules during the pionic cascade. For this purpose, we measured the electronic X rays correlated with pionic X rays. The experiments are performed at the pi-mu-channel of KEK-PS. We constructed a new measurement system. This apparatus was improved for efficient measurements of correlated events. We used Zn, Mo and Sn metals and their oxides as the targets, and examined correlation between electronic and pionic X rays.

1P18: Measurement of hyperfine field in mavicyanin by perturbed angular correlation of γ-rays

Hashimoto, T.[1], Yokoyama, A.[2], Kataoka, K.[2], Takata, M.[1], Ohkubo, Y.[3] (Graduate School of Natural Science and Technology, Kanazawa Univ.[1], Department of Chemistry, Faculty of Science, Kanazawa Univ.[2], Research Reactor Institute, Kyoto Univ.[3])

Measurement of perturbed angular correlation of γ-rays (PAC) is unique method to investigate a hyperfine field in a molecule in solution based on nuclear electric quadrupole interaction. Taking advantage of the method, it is interesting to measure the hyperfine field at a metal site of biological molecule in an aqueous solution as functioning in life. In this study, the structure of metal site of Mavicyanin, a protein molecule with a site of copper, was investigated by using time-differential perturbed angular correlation of γ-rays of ^{117}Cd. The time dependences of coincidence counts of the 90-344 keV cascade γ rays for the sample containing ^{117}Cd were taken using a measurement system consisting of standard fast-slow electronic modules and four BaF_2 scintillation detectors. Compared with the electric field gradient reported on Stellacyanin, which has a molecule structure similar to Mavicyanin, the obtained results in this study seem to be reasonable.

1P19: TDPAC and Mössbauer spectroscopic studies on Fe/Mo multilayers

Murakami, Y.,[a] Ohkubo, Y.,[b] Sakamoto, Y.,[c] Fuse, D.,[c] Ono, T.,[c] Kitao, S.,[b] Seto, M.,[b] Tanigaki, M.,[b] Saito, T.,[d] Nasu, S.,[c] Kawase, Y.[b] ([a] Grad. Sch. Sci., Kyoto Univ. [b]Res. Reactor Inst., Kyoto Univ. [c]Grad. Sch. Eng. Sci., Osaka Univ. [d]Radioisotope Res. Center, Osaka Univ.)

Knowledge of the structure and the magnetic properties at the interface of artificial magnetic multilayers is very important for understanding interlayer exchange coupling and giant magnetoresistance (GMR) of the multilayers. This time, we performed ^{57}Fe-Mössbauer and ^{99}Tc-TDPAC measurements on Fe/Mo multilayers. Mössbauer spectroscopy and TDPAC are known to be powerful techniques for investigating the local electronic states of materials through the hyperfine interaction parameters for probe nuclei. Mössbauer spectroscopy and TDPAC were used to measure the local magnetic field in the Fe layers and in the Mo layers of Fe/Mo, respectively. The results show that the magnetic structure in Fe and Mo layers varies with the Mo layer thickness. We consider that the variation reflects the interlayer coupling between two Fe layers.

1P20: PAC studies on ^{111}Cd and ^{117}In in pyrochlore $Cd_2Nb_2O_7$

Ohkubo, Y. (Res. Reactor Inst., Kyoto Univ.), Murakami, Y. (Grad. School Sci., Kyoto Univ.), Saito, T. (Radioisotope Res. Center, Osaka Univ.), Yokoyama A. (Fac. Sci., Kanazawa Univ.)

The nuclear-electric-quadrupole interactions at ^{111}Cd and ^{117}In nuclei arising from ^{111m}Cd and ^{117}Cd, respectively, chemically introduced in pyrochlore ferroelectric $Cd_2Nb_2O_7$ (T_C = 196 K) were studied at 77, 290, and 1073 K, using time-differential perturbed-angular-correlation (PAC) technique. At temperatures above T_C, there is one type of Cd sites. However, at 77 K (below T_C) there are two types of Cd sites. The ratio of the electric quadrupole frequency of ^{117}In to that of ^{111}Cd is anomalously deviated from the value expected from the purely-ionic In and Cd ions.

1P21: Development of neutron in-beam Mössbauer spectroscopy

Kubo, M. K.[a], Kobayashi, Y.[b], Saito, T.[c], Yamada, Y.[c] Sakai, Y.[d], Shoji, H.[e], Yonezawa, C.[f], Matsue, H.[f], Sato, W.[b], Nakada, M.[f] ([a]ICU, [b]RIKEN, [c]Sci. Univ. Tokyo, [d]Daido Inst. Tech., [e]Tokyo Metro. Univ., [f]JAERI)

We have been developing a neutron in-beam Mössbauer spectroscopy by using thermal and cold neutron beams at the research reactor JRR-3M in JAERI. In-beam Mössbauer spectrometry is a promising tool for detection and characterization of trace exotic chemical species produced via ion implantation under special conditions such as low temperature, high pressure, *etc.* We selected the ^{56}Fe(n,γ)^{57}Fe reaction for the first candidate and succeeded in obtaining Mössbauer spectra of pure iron and stainless steel by using a parallel plate avalanche counter (PPAC). In order to apply this method to various chemical compounds containing less iron, we need improve the signal to noise ratio that is not satisfactory so far. We installed a Li metal neutron guard and a specially made Pb shield and are planning to reduce the size of the PPAC inside which high energy gamma-rays could produce noise electrons.

1P22: Mössbauer spectroscopy of ^{133}Cs using ^{133}Xe-implanted C_{60} and C_{70} fullerenes

Muramatsu, H.,[a] Shimomura, H.,[a] Watanabe, S.,[b] Ishioka, N.S.,[b] Kume, T.[b] ([a]Shinshu Univ., [b]JAERI)

The Mössbauer emission spectra have been measured using ^{133}Xe-implanted C_{60} and C_{70} fullerenes. Fullerene targets, which were prepared by vacuum evaporation of C_{60} or C_{70} on a Ni foil, were bombarded by ^{133}Xe ions with kinetic energies of 30-38 keV by using an electromagnetic isotope separator at doses raging from 1×10^{12} to 1×10^{14} cm^{-2}. The measurements of Mössbauer effect in ^{133}Cs were carried out at 4.2K by using two different types of sample, "as implanted" right after the implantation and an insoluble portion after the dissolution of the target in *o*-dichlorobenzene. The absorption intensities for both samples were extremely small, which means that the recoilless fractions in both samples should have extremely small values and implanted atoms would be bound to the lattice with considerably weak forces or have enough space to vibrate with a large scale. Both isomer shifts were close to the value of +0.05 mm/s (w.r.t. CsCl absorber) in some compounds such as $CsMnF_3$, which corresponds to ^{133}Cs in a site with the lowest electron density observed up to now in any ^{133}Cs Mössbauer measurement, where Cs is considered to be in the $5s^25p^66s^0$ electron configuration.

1P23: Synthesis and photo magnetic properties for UV, Vis or IR-driven spin transition compounds

Shigeyoshi, Y.,[a] Hyami, S.,[a] Inoue, K.,[b] Maeda, Y.,[a] ([a]Kyushu Univ., [b]IMS)

The Fe(II) compounds $[Fe(aza)_2(NCS)_2]$ (1) and $[Fe(sty)_2(NCS)_2]$ (2) have been synthesized and their magnetic susceptibilities have been measured. Compounds 1 and 2 are spin-crossover compounds with spin transition temperature $T_{1/2}$ = 180 K (for compound 1) and $T_{1/2}$ = 210 K (for compound 2), and do

not show a hysteresis loops around the spin transition temperature. The magnetic susceptibilities and Mössbauer spectra after illumination at 550 nm prove that compound **1** exhibits the LIESST effect. Compound **2** also exhibits the LIESST effect after illumination at 550 nm and even at 800 nm. The ligands aza and sty in the compounds **1** and **2** have photo-isomerization moieties and are expected to occur the LD-LISC (Light-Driven Ligand Isomerization Spin Change). The effect of illumination at 350 nm has been measured for the compounds **1**-PMMA and **2**-PMMA diluted in polymer matrix PMMA (poly methyl methacrylate).

1P24: Mossbauer spectroscopic study of lanthanoid-iron complex

Matsumoto, K., Katada, M. (Graduate School Science, Tokyo Metropolitan Univercity)

We have reported Mössbauer spectroscopic studies of $Ln[Fe(CN)_6] \cdot nH_2O$ (Ln=La-Lu) and their complexes with 1,8-Naphthyridine , 2,2'-Bipyridyl , 1,10-Phenanthroline and 2,2';6'2''-Terpyridine . In this study , we prepared new complex by using $La(phen)_2Cl_3$, $Na_3[Fe(CN)_5L] \cdot nH_2O$ (L= n-Butylamine , n-Octylamine) as a starting material . The obtained complex were characteried by means of Mössbauer spectroscopy .

1P25: Mössbauer spectroscopic studies of hexacyanoferrate(III) complexes $[Cu(L)]_3[Fe(CN)_6]_2 \cdot nH_2O$

Suzuki, N., Katada, M. (Graduate School of Science, Tokyo Metropolitan Univ.)

Some complexes of the type $[Cu(L)]_3[Fe(CN)_6]_2 \cdot nH_2O$ (L=ethylendiamine(en)[1,2], diethylentriamine(dien)[3], 2,2'-bipyridine(bpy)[4], 2,2':6',2''-terpyridine(terpy)[5]) have been prepared and characterized by means of XRD, TG-DTA and Mössbauer spectroscopy. Mössbauer parameters at room temperature, isomer shift (IS) and quadrupole splitting (QS) were -0.10~-0.14 mms^{-1} and 0.48~0.74 mms^{-1},respectively. The QS value of [5] were 1.41mms^{-1}. The value of IS are typical low spin state iron(III). The value of QS for [5] is contributed to lowering symmetry of Fe(III) site.

1P26: Mössbauer spectra of iron complexes of tetrahydroxy-1,4-benzoquinone

Tooyama, Y., Matsuoka, N., Fujii, S., Sakai, H. (Fac.of Sci. and Eng., Konan Univ.)

Iron complexes of tetrahydroxy-1,4-benzoquinone were characterized from Mössbauer and IR spectra, and XRD patterns. Interestingly, the complex prepared from iron(II) sulfate indicates the same Mössbauer spectrum as one prepared from iron(III) sulfate, consisting of iron(II) and iron(III) species. Furthermore, the XRD patterns and IR spectra for both complexes resemble each other. Tetrahydroxy-1,4-benzoquinone acts likely as reductant at high temperature, as well as 2,5-dichloro-3,6-dihydroxy-1,4-benzoquinone.

1P27: Mössbauer spectra of the dinuclear iron complexes with the bridging dihydroxybenzoquinone ligand

Matsuoka, N., Tooyama, Y., Fujii, S., Sakai, H. (Fac.of Sci. and Eng., Konan Univ.)

Dinuclear bipyridil-iron complexes with the bridging dihydroxybenzoquinone ligand were prepared and characterized from Mössbauer, ESR and IR spectra, and XRD patterns. The iron content is analyzed to be 12.7% by using the atomic absorption spectroscopy, suggesting the complex to be the formula $[Fe_2(bpy)_4(DHBQ)]$. The Mössbauer spectrum indicates that the iron ions are divalent low spin state in the complex. The complex exhibits the strong ESR signal near g=2 in the solid state, due probably to the bridging (DHBQ) trivalent anion.

1P28: ^{121}Sb Mössbauer spectra for some organometallic complexes having Zintl anion

Kitadai, K., Takahashi, M., Takeda, M. (Department of Chemistry, Faculty of Science, Toho Univ.)

^{121}Sb Mössbauer spectra for $[Sb_7]^{3-}$, $[Sb_7M(CO)_3]^{3-}$ (M = Cr, Mo), $[Sb_7Ni_3(CO)_3]^{3-}$ were measured at 12 K. Two antimony sites are observed in all compounds. The values of isomer shift (δ), relative to InSb, indicate that the electrons in the Sb_7^{3-} are slightly delocalized over the Sb_7^{3-} anion (δ = –2.56, –2.57, –2.77 and –2.02 mm s^{-1}, respectively). The negative values of e^2qQ were observed for Sb sites that coordinate to transition metal in $[Sb_7M(CO)_3]^{3-}$ (M = Cr, Mo), while corresponding Sb site in non–coordinate Sb_7^{3-} showed rather large positive e^2qQ values. This suggests that some electrons were transferred from Sb atom to M atom in the organometallic compounds. The Mössbauer parameters for $[Sb_7Ni_3(CO)_3]^{3-}$ are much different from those of other organometallic compounds due to the rearrangement of Sb atoms in Sb_7^{3-} unit.

1P29: ^{99}Ru Mössbauer spectroscopic studies of $CaRuO_3$

Kobayashi, Y., Okada, T., Haba, H., Jimbo, S.[1], Taniguchi, T.[1], Noro, Y.[2], Nagata, Y.[1] (RIKEN, [1]College of Science and Engineering, Aoyama Gakuin University, [2]Kawazoe Future Technology)

Ternary ruthenates, $MRuO_3$ (M = Sr and Ca), have nearly cubic and slightly distorted cubic perovskite structure, respectively. Although both ruthenates show metal-like conductivity, their magnetic behavior are quite different: $SrRuO_3$ is well-known to be an itinerant ferromagnetic metal with the Curie temperature T_C~ 160 K, whereas the magnetic ground state of $CaRuO_3$ is more controversial. Recent papers have suggested that magnetic properties of $CaRuO_3$ are exchange enhanced paramagnetic, antiferromagnetic, weak ferromagnetic, and/or a spin-glass type. In this paper, we prepared single crystals of $CaRuO_3$ grown by the flux method using $CaCl_2$ as a flux, and measured ^{99}Ru Mössbauer spectroscopy of the samples. The magnetic ground state of $CaRuO_3$ will be discussed compared to the magnetic property of $SrRuO_3$.

1P30: Photoinduced magnetization for cyano-bridged 3d-4f hetero-bimetallic assembly

Einaga, Y.,[a] Li, G.-M.,[a] Akitsu, T.,[a] Sato, O.[b] ([a]Keio Univ., [b]KAST)

Photo-induced magnetic and/or optical properties have attracted much attention since photo-control of the magnetic and optical properties remains a challenging topic for material science in view of the possible implementation in optical and memory devices. It is noted that almost all previous reports were focused on Prussian blue analogues or transition metal complexes. However, It is difficult for Prussian blue analogues to be crystallized so that the proposed structures could not be verified by X-ray crystallographic analysis. In contrast, various cyano-bridged 3d-4f hetero-bimetallic assemblies have been synthesized and structurally characterized driven by the interesting in molecular magnetism recently. Keeping the similarities of the two types of complexes in mind, we studied the effects of magnetic properties by light stimulation on the cyano-bridged 3d-4f hetero-bimetallic assemblies. As a result, a significant photoinduced magnetization was observed for $Nd(DMF)_4(H_2O)_3(\mu\text{-}CN)Fe(CN)_5 \cdot H_2O$ (DMF = *N, N* –dimethylformamide) (1) in a given range of temperature.

1P31: The chemical states of heavy metals in estuarine and tideland sediments

Kataoka, M.,[a] Matsuo, M.[b] ([a]Graduate School of Sci., The Univ. of Tokyo, [b]Graduate School of Arts and Sci., The Univ. of Tokyo)

We collected sediments vertically in Yatsu tideland and Arakawa-river, which are located at the east part of Tokyo. Yatsu tideland was selected as a non-polluted area and Arakawa-river was assumed to be polluted with the human activities. Chemical states of

iron were investigated by Mössbauer spectroscopy. In the vertical distribution of iron species, maximum pyrite distribution was found in the middle layer. The distribution of pyrite was similar in the both areas, suggesting that pyrite formation in the sediment is not affected by anthropogenic effects. Chemical states of manganese and zinc in the sediments were investigated by X-ray absorption fine structure (XAFS). From the shape of each manganese spectrum, MnS was estimated to be rich in the surface layer. Zinc existed as divalent components in all layers. However, the counter anion changed from O to S with increasing depth. Thus, the distribution pattern of sulfides in the sediments was different according to the difference of the heavy metals of the sulfides.

1P32: Carbon-implanted polyethylene studied by a pulsed slow-positron beam

Djourelov, N.[a], Suzuki, T.[a], He, C.Q.[a], Ito, Y.[b], Hamada, E.[c], Kondo K.[a] ([a]KEK, [b]RCNT, [c]Inst. for Environmental Sciences)

The pulsed slow-positron beam technique was applied to study the structural changes in ultra-high molecular-weight polyethylene after carbon implantation at three implantation doses. The influence of additional gamma-irradiation on the sample's structure was studied. A three-layer model was used in order the experimental results to be explained. An increase in the number of pores and a decrease in their sizes were observed for a subsurface layer. A low-porosity layer next to the subsurface layer was detected. The position of the former, at the lowest carbon concentration, was in good agreement with the calculated one by TRIM. In the case of higher carbon concentrations significant discrepancy of the positions was observed.

1P33: Coincidence Doppler broadening spectroscopy (CDBS) in hydro-carbon

Suzuki T.[a], Djourelov N.[a], Ito Y.[a], Kondo K.[a], Ito Y.[b] ([a]KEK, [b]RCNT)

Positron annihilation (PA) has been used widely to study electronic states in various field: Physics, Engineering, Chemustry. In this research, PA was applied to study PA in Hydro-Carbon. Coincidence Doppler broadening spectroscopy (CDBS), which uses two Ge detectors, can improve SN ratio by three orders of magnitude. In this way, PA with higher momentum electrons can be detected. The comparison between PA in methane and ethylene showed the effect of π and σ electrons. Methane has only σ electrons and ethylene has π and σ electrons. Three samples: diphenyl, anthracene, and naphthalene, have benzene ring in their chemical structure. Although PA in threes samples have effects of Ps formation, other effect was not observed. This shows that positrons annihilate with the similar structure of electrons (π and σ electrons).

1P34: Dynamic behavior of energetic hydrogen isotope in SiC

Onishi,Y.,[a] Nunome, T.,[b] Kodama, H.,[b] Oya, Y.,[c] Okuno, K.,[b] Tanaka, S.[d], Makide,Y.[c] ([a]Fac. of Sci., Tokyo Univ. of Sci., [b]Radiochem. Res. Lab., Shizuoka Univ., [c]Radioisotope Center, The Univ. of Tokyo, [d]Grad. School of Eng., The Univ. of Tokyo)

Silicon carbide (SiC) is considered to be a candidate material for the first wall of future fusion reactors. However, the behavior of hydrogen isotope in SiC was not clarified sufficiently. In this study, the dynamic behavior and chemical state of energetic hydrogen isotope into SiC have been studied by means of thermal desorption spectroscopy (TDS) and X-ray Photoelectron spectroscopy (XPS). The 1.0 keV deuterium ions were implanted into β-SiC sample with keeping the temperature between 173K and 1073K. Two desorption peaks were observed at 790K and 910K by TDS. In the XPS analysis, the C 1s and Si 2p peaks were shifted to higher and lower energy side by the deuterium ion irradiation. These facts indicate that deuterium ions were bound to Si and C, respectively. From XPS and TDS analyses, the deuterium desorption processes at 790K and 910K correspond to the desorption of deuterium bound to Si and C, respectively.

1P35: Studies on hot atom chemical behavior of energetic ions in solids (IV) -Chemical behavior of energetic deuterium implanted in boron thin film-

Yoshikawa, A., Kodama, H., Oyaidzu, M., Oya, Y.,[a] Matsuyama, M.,[b] Sagara, A.,[c] Noda, N.,[c] Okuno, K., (Radiochem. Res. Lab., Shizuoka Univ. RI center, Univ. of Tokyo.,[a] HIRC., Toyama Univ.,[b] NIFS[c])

In D-T fusion reactors, boron coating film on plasma facing materials (PFMs), boronization, could be a promising method to reduce impurities in D-T plasma. Though it is important to elucidate hot atom chemical behavior of energetic tritium irradiated into the boron thin film from the viewpoint of fusion reactor safety, little has been studied yet. Therefore, we studied chemical behavior of energetic deuterium implanted in boron thin films, which were prepared by PCVD using $B_{10}H_{14}$ diluted with He gas. For the boron thin films with 87 % purity of boron, energetic D_2^+ implantation was performed at temperatures from 173 K to 873 K, and its thermal desorption behavior was measured. In the Thermal Desorption Spectroscopy (TDS) spectrum of D_2, two peaks were observed at about 450 K and 650 K. It was also found that with increasing implantation temperature, the peak at 650 K reduced more quickly than that at 450 K. Based on these TDS results, we'll discuss in details hot atom chemical behavior of energetic deuterium implanted boron thin film.

1P36: Studies on hot atom chemical behavior of energetic ions in solids (V) —Chemical behavior of energetic deuterium ions in carbon material—

Nunome, T.[1], Kimura, H.[1], Sasaki, M.[1], Morimoto, Y.[1], Okuno, K.[1], Takahasi, K.[2], Sakamoto, K.[2], Imai, T.[2](Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ.[1], JAERI[2])

A torus diamond window installed in the radio frequency heating system of fusion reactors is in the circumstance of energetic tritium, deuterium and helium and radio activated dust from plasma. It is, therefore, important to elucidate the chemical structure changes of the diamond and existing states of the irradiated ions from the viewpoint of optical characteristics of the window and safety issue of the fusion reactors. In this study, the structural change of diamonds induced by energetic deuterium and helium ions and their existing states was investigated by X-ray Photoelectron Spectroscopy and Thermal Desorption Spectroscopy. In both results from the irradiation of deuterium and helium ions, C1s peak shifted toward lower binding energy side with increasing their fluence. C1s peak resulted from the deuterium irradiation, however, was located at higher energy side than that from the helium irradiation. This result indicated that the C-D bonding was formed by the deuterium irradiation and then the diamond could change to amorphous carbon due to formation of C-D bond.

1P37: Experimental stations for high-resolution photoemission spectroscopy and magnetic circular dichroism spectroscopy in soft x-ray beamline BL23SU at SPring-8

Muramatsu, Y.,[a] Okamoto, J.,[a] Mamiya, K.,[a] Fujimori, S.,[a] Okane, T.,[a] Saito, Y.,[a] Fujimori, A.[a,b] ([a]JAERI, [b]Tokyo Univ.)

Experimental stations for high-resolution photoemission spectroscopy (HRPES) and soft x-ray magnetic circular dichroism (MCD) spectroscopy, which have been installed in radioisotope (RI) facility at beamline BL23SU/SPring-8, are presented. The HRPES station provides electronic structure information near Fermi level of functional materials, and the MCD station provides the spin/orbital-angular-momentum information of magnetic materials.

1P38: Dissolution behavior of uranium oxide in a

different composition of the HNO_3-TBP complex

Shimizu, R., Sawada, K., Tsushima, S., Enokida, Y., Yamamoto, I. (Nagoya University)

HNO_3-TBP complex is obtained as organic solution after contacting tri-*n*-butyl phosphate (TBP) and HNO_3. This complex dissolves metal oxides and extracts as $M(NO_3)_m \cdot n$TBP. A 20 cm^3 of TBP (Wako Chemicals) was contacted with different volumes of concentrated HNO_3 (Wako Chemicals) by shaking vigorously in a glass tube to prepare HNO_3-TBP complex. The concentration of HNO_3 in prepared HNO_3-TBP complexes was determined by a titration with NaOH solution, after HNO_3 in the complex was back extracted into the aqueous solution. The total content of H_2O was determined by Karl Fischer titration method. Electrical conductivity and viscosity of the complexes were measured by MPC227 (Mettler Toledo Co.) and VM-1 (CBC Materials) at 298 K. Molecular ratio of HNO_3 to H_2O and electrical conductivity as a function of acidities of HNO_3-TBP complex shows clearly that there may be two different chemical structures on a boundary 3 M. UO_2 powder obtained by grinding mechanically UO_2 pellet was dissolved in the different composition of the HNO_3-TBP complex to investigate dissolution behavior.

1P39: Separation of actinides in solid samples by supercritical carbon dioxide leaching method; Separation behavior of uranium and thorium with HNO_3-TBP complex

Meguro, Y.,[a] Imai, T.,[a] Tomioka,[a] O., Yoshida, Z.,[a] Nakashima, M.,[a] Tsushima, S.,[b] Wada, R.[c] ([a]JAERI, [b]Nagoya Univ., [c]KOBELCO)

Supercritical CO_2 fluid leaching (SFL) method using supercritical carbon dioxide containing HNO_3 - tri-n-butyl phosphate (TBP) complex as a reactant has been developed for removal and recovery of uranium from solid samples. The SFL procedure consists of the complexation process of uranium with the supercritical CO_2 containing the HNO_3-TBP complex at 60 °C and 15 MPa and the dissolution process of the uranium complex with a flow of supercritical CO_2 (8 mL/min) for 75 min at 60 °C and 20 MPa, and these two processes are regarded as one cycle SFL. Two cycles SFL using the supercritical carbon dioxide containing 2 mL HNO_3-TBP complex was applied to the separation of uranium from 5 g synthetic ash sample contaminated by 100 mg uranium containing ca. 1 pg thorium as a daughter nuclide of ^{238}U. More than 99% of uranium and 98% of thorium were separated form the ash. Separation efficiency of uranium was slightly larger than that of thorium.

1P40: Oxidation-reduction of uranium ion by means of flow-coulometry with column electrode

Aoyagi, H.[a], Kitatsuji, Y.[a], Kimura, T.[a], Yoshida, Z.[a], Kihara, S.[b] ([a]JAERI, [b]Kyoto Inst. of Technol.)

The oxidation-reduction of U(IV) / U(VI) couple was studied by means of flow-coulometry with the column electrode using glassy carbon fiber as a working electrode. Two-electrons oxidation waves were observed for the oxidation of U(IV), and the oxidation waves of lower acidic solution were shifted to more negative potential. A one-step oxidation wave of low reversibility was obtained for U(IV) in 10 M $HClO_4$; a two-steps oxidation wave was observed in the coulopotentiograms for the U(IV) in 2.0 – 5.0 M $HClO_4$; a one-step two-electrons oxidation wave was observed for the U(IV) in lower concentration of $HClO_4$. It is suggested that there are at least two reaction passes for the oxidation of U(IV), and that other U(IV) species different from U^{4+} is concerned in the reaction of the first oxidation wave.

1P41: XAFS and ^{237}Np Mössbauer spectroscopic studies of neptunyl hydroxides

Nakada, M., Otobe, H., Akabori, M., Yamashita, T., Minato, K., Motohashi, H. (JAERI)

It is difficult to study actinide elements for high radioactivities. ^{237}Np compounds must be packed and sealed in sample containers for measurement, as ^{237}Np is an α-ray emitter. We have been studying chemical properties of some neptunyl compounds sealed in sample containers using ^{237}Np Mössbauer spectroscopy. XAFS measurements also can be used sealed samples. We attempted the XAFS measurements of neptunium compounds. Several mg of neptunium compounds, $^{237}NpO_2$, $^{237}NpO_2(OH)$ and $^{237}NpO_2(OH)_2$, and carbon powder were pressed into small discs of 7 mm in diameter, respectively. The discs were put into PET containers and sealed by resin (stycast 1266). XAFS measurements were conducted at KEK PF on the BL-27B endstation. All the XAFS spectra were collected at the Np L_{III}-edge in transmission mode at room temperature using a Si(111) double crystal monochromator. The white line position of $NpO_2(OH)_2$ was shifted by a few eV with respect to $NpO_2(OH)$ on XANES spectra. There was a small shoulder above the white line maximum in the spectra of the neptunyl samples.

1P42: Reduction of neptunium(V) on magnetite surface

Kitamura, A.[a], Nakata, K.[b], Tanaka, S.[b], Tomura, T.[c], Kamei, G.[a] ([a]JNC, [b]Univ. of Tokyo, [c]Inspection and Development Co.)

Redox reactions between neptunium(V) (Np(V)) and magnetite ($Fe(II)_1Fe(III)_2O_4$) surface were investigated. A batch method was applied to the experiment. High-pure magnetite (99.9 %, obtained from Rare Metallic Co.) and a 0.1 M NaCl were mixed in a polypropylene tube, and pH, redox potential and concentration of dissolved neptunium were measured as a function of shaking time (from 1 hour to 7 days), temperature (25 °C and 45 °C) and liquid/solid ratio (20, 50 and 100 $ml.g^{-1}$). It was observed that the concentration of dissolved neptunium was reduced rapidly within a day, due to the reducing of Np(V) to Np(IV) and the precipitation of Np(IV). This result was shown typically when the magnetite/solution ratio and the temperature were high. The rate constant of the redox reaction and the activation energy for the rate constant were preliminarily obtained. It was suggested that the redox reaction was promoted by not only Fe(II) on magnetite surface but also Fe(II) inside the magnetite.

1P43: Effects of dry density and ionic strength on transport number of sodium ions in compacted Na-montmorillonite

Akagi, Y.,[a] Higashihara, T.,[a] Kinoshita, K.,[b] Sato, S.,[a] Kozaki, T.[a] ([a]Graduate School of Eng., Hokkaido Univ.,[b]Mitsubishi Electric Corp.)

Our studies are to discuss the effects of dry density and ionic strength on transport numbers of sodium ions in compacted Na-montmorillonite saturated with $NaClO_4$ solutions. The transport numbers of sodium ions were from 0.67 to 0.85 in the region of dry densities from 1.0 to 1.6 Mg m^{-3}, while were from 0.42 to 0.77 in the region of ionic strength from 0.1 to 0.5 mol l^{-1}.

1P44: Migration behavior of Np(V) in water-saturated, compacted Na-montmorillonite under an electric potential gradient

Higashihara, T.,[a] Akagi, Y.,[a] Sato, S.,[a] Kozaki, T.,[a] Mitsugashira, T.,[b] Hara, M.,[b] Suzuki, Y.[b] ([a]Div. of Quantum Energy Eng., Graduate School of Eng., Hokkaido Univ., [b]Oarai Branch, Institute for Materials Res., Tohoku Univ.)

As a part of nuclear waste disposal, $^{239}Np(V)$ was migrated in water-saturated, compacted montmorillonite under an electric potential gradient. The compacted Na-montmorillonite columns saturated with 0.1 M $NaClO_4$ and Na_2CO_3 solutions were prepared at the dry density of 1.0 Mg m^{-3}. Under an electric potential gradient, Np(V) tracers dissolved in 0.1 M $NaClO_4$ as cations, and 0.1 M Na_2CO_3 as anions were migrated in the compacted montmorillonite for 20 h. After the polarization, the concentration

profiles of Np(V) in the montmorillonite were obtained. Not only the cation tracer of Np(V) but also the anion one were not migrated in the compacted montmorillonite, though 22Na+ and 36Cl- were appreciably migrated toward to cathode and anode, respectively. These results suggest that Np(V) dissolved in the water contained in the compacted montmorillonite sorbed strongly on the clay particles differing from results by a computer code.

1P45: Adsorption behavior of trivalent lanthanides and actinides on tertiary pyridine resin in nitric acid solution

Itoh, K.,[1] Otsuka, M.,[1] Suzuki, T.,[2] Ikeda, A.,[2] Ohtake, K.,[2] Aida, M.,[2] Fujii, Y.,[2] Ozawa, M.,[2,3] Hara, M.,[4] Mitsugashira, T.[4] (Shibaura Inst. Tech.[1], Tokyo Tech.[2], JNC[3], Tohoku Univ.[4])

The group and mutual separations of the trivalent lanthanides and actinides have been studied by means of the elution chromatography using the columns packed with the tertiary pyridine type resin embedded in silica beads. In the present work, the adsorption behavior of trivalent lanthanides and actinides on pyridine resin in the nitric acid and/or nitric acid and methanol mixed solvent were investigated, and were compared with the hydrochloric acid system. It was confirmed that the group separation of lanthanides and actinides are difficult since the americium and curium ions are eluted between cerium and neodymium ions, and the separation factors of elements among same group are higher than the hydrochloric acid system. These results show that the adsorption mechanism is different between the nitric acid system and the hydrochloric acid system.

1P46: The formation constant between trivalent f-block elements and thiocyanate ion

Mori, T., Suganuma, H., Yanaga, M. (Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ.), Satoh, I. (IMR, Tohoku Univ.)

The formation constants of thiocyanate complexes of Eu(III), ^{152,154}Eu, and Am(III), ^{241}Am, in trace concentrations were investigated in mixed solvent (CH_3OH + H_2O) solutions. The formation constant was obtained by a back-extraction technique as follows. Aliquots of 10.0 ml of 1.0 M (H, Na)(SCN, ClO_4) + x M LiCl in mixed (CH_3OH + H_2O) solvent solution were placed in a plastic vial. An aliquot of 5.0 ml of the organic phase, bis(2-ethyl-hexyl)hydrogenphosphate (HDEHP), containing the radionuclied was added to the vial and the mixture was shaken for more than 1 h at 298 K. The obtained results showed that the formation constants of thiocyanate complexes of Eu(III) and Am(III) were approximately same values in 1.0 M (H, Na)(SCN, ClO_4) + 5.0 M LiCl aqueous solution but the formation constant of thiocyanate complex of Am(III) was considerably larger than that of Eu(III) in 1.0 M(H, Na)(SCN, ClO_4) mixed solvent solution (X_{MeOH} = 0.60).

1P47: Solvent extraction of f-block element ions by thiacalix[4]arenes and their oxides

Matsuyama, K., Suganuma, H., Yanaga, M., Satoh, I.[1] (Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ., IMR, Tohoku Univ.[1])

It is expected that thiacalix[4]arenes and their oxides (sulfinyl- and sulfonylcalix[4]arenes) have the ability of selective complexation for metal ion, based on HSAB principle. We synthesised thiacalix[4]arenes and their oxides from two kinds of *p*-alkylphenol (*p-tert*-butyl- and *p-tert*-octylphenol). The extraction behavior of ^{152,154}Eu(III) and ^{241}Am(III) in region of 3.2<pH<6.1 was investigated using those compounds dissolved into chloroform. The extraction behaviors of ^{152,154}Eu(III) and ^{241}Am(III), except *p-tert*-butylsulfinylcalix[4]arene and *p-tert*-butylsulfonylcalix[4]arene, were nearly same, but the extraction by *p-tert*-butylsufinylcalix[4]arene and *p-tert*-butylsulfinylcalix[4]arene showed somewhat difference between both elements.

1P48: Synthesis and structural determination of La complex with tridentate ligand *N,N'*-dimethyl-*N,N'*-diphenyl-2,6-pyridinedicarboxy amide (DMDPhPDA)

Shimada, A.,[a,b] Yaita, T.,[b] Kimura, T.,[b] Okuno, K.[a] ([a]Graduate School of Sci. and Engineering., Shizuoka Univ., [b]JAERI)

N,N'-dimethyl-*N,N'*-diphenyl-2,6-pyridinedicarboxyamide (DMDPhPDA) is one of the promising ligand for separation of trivalent actinides from trivalent lanthanides in high level nuclear waste. We have studied the extraction properties of trivalent lanthanides with the DMDPhPDA. In this work, the single crystal of La-DMDPhPDA complex, $[La(NO_3)_3(DMDPhPDA)_2]$, was synthesized and its structure was determined by X-ray diffraction method in order to elucidate extraction mechanism for lanthanides with DMDPhPDA from the view-point of structural study. The $[La(NO_3)_3(DMDPhPDA)_2]$ shows that La takes a 12 hold coordinated ion structure with three bidentate nitrate ions and two tridentate DMDPhPDA ligands. Water molecules are not found in the coordination structure. The bond lengths of La-O (carbonyl group) [ave. 2.6755 Å] are shorter than those of La-N (pyridyl group) [2.782(3) Å]. These bond lengths are nearly equal to those of the La complexes with 12 coordination structure including oxygen and nitrogen as donor atoms.

2P01: Determination of extractable organofluorine (EOF) in fish samples from Matsuyama using instrumental neutron activation analysis

Kawano, M., Nakatsu, T., Wakimoto, T. (Faculty of Agriculture, Ehime University)

A new analytical method for extractable organofluorine (EOF) in environmental sample using instrumental neutron activation techniqu has been developed. Organofluorine compounds have been used in industry and daily life. EOF in environmental sample was extracted with organic solvent by using sohxlet extractor. Activation analysis was carried out by irradiation condition of neutron flux at a rate of 3.5 x 10^{13} n/cm^2 · sec for thirty seconds using the reactor JRR-4 , Japan Atomic Energy Research Institute (JAERI). The gamma-ray from ^{20}F was measured by gamma-ray spectrometry. EOF concentrations in extract samples of freshwater and seawater fishes collected in Matsuyama were determined by the technique. However, EOF in the samples investigated was not detected. It may suggest that the samples were contaminated by EOF less than 10 μg/g (on fat weight basis). It is necessary to investigate on the other fraction such as water soluble one of the same sample as well as the other environmental samples.

2P02: Neutron activation analysis of Selenium in human toe nails

Seki, R., Usui, T., Matsuhiro T., Ohno, S., (Department of Chemistry, University of Tsukuba)

Selenium concentration in human toe nails has been measured by instrumental neutron activation analysis. In the analysis we used short-lived ^{77m}Se (half-life: 17.36 s) generated by the ^{76}Se(n, γ)^{77m}Se reaction. The analysis was performed at JAERI, JRR-3 reactor, using a pneumatic tube, PN-3 (ϕ_{th}=1.5×10^{13}cm^{-2}s^{-1}). Each sample was irradiated for 20 seconds then transferred and counted for 30 seconds with Germanium detector 8 seconds after irradiation. The overall average toe nail specimens found in this study was found to be 1.14±0.14 ppm and similar to Canadian subjects. There is no significant difference in this study between sexes and between smoker and non-smoker.

2P03: Determination of the concentrations of trace elements in organs and tissues of zinc-deficient mice by means of INAA and PIXE analysis

Ogi, T., Kajita, Y., Kinugawa, N., Ohyama, T., Noguchi, M.,

Suganuma, H., Yanaga, M. (Fac. of Sci., Shizuoka Univ.)

Effects of zinc deficiency on metabolism of trace elements in liver of mice were investigated. Eight-week old male mice of ICR strain were divided into two groups; one was fed with zinc-deficient diet (<1 μg/g Zn), the other with control diet (30 μg/g Zn) at libitum for 3 weeks. After the treatment periods, their livers were removed. Two types of experiments were performed. In the first experiment, the liver samples homogenized with HEPES buffer were centrifuged under differential conditions to separate into nulear, mitochondrial, lysosomal, microsomal and cytosolic fractions. Each fraction was freeze-dried and subject to an instrumental neutron activation analysis (INAA). In the second experiment, SDS-PAGE electrophoresis was performed for cytosolic fraction. Then, the electrophoresis gel was cut into small pieces. The elemental concentrations in each piece were determined by a particle induced X-ray emission (PIXE) analysis. Zinc concentration in each hepatocyte component was different between Zn-def. and control mice, although no significant differences were recognized in whole liver analysis. In the electrophoresis result, specific protein due to zinc deficiency was not found.

2P04: The analysis of cadmium-polluted rice by using multiple gamma-ray detecting method

Toh ,Y., Oshima, M., Hatsukawa, Y., Koizumi, M., Osa, A., Kimura, A., Jun, G. (JAERI)

Since Japan existed in the volcano zone and mining development has been performed positively, it is in the tendency for the cadmium concentration to be high, and the cadmium concentration of agricultural products becomes comparatively high. The issue on cadmium concerns all foods, and especially, rice is the most important staple food in Japan. By applying the multiple gamma ray detecting method to PGNAA, the influence from nuclei which emits only one prompt gamma ray simultaneously can be eliminated. If it is possible to remove the influence of hydrogen contained in rice by using this new method, the detection limit of cadmium is improved significantly. In this research, the detection limit of cadmium contained in the rice by Prompt Gamma Neutron Activation Analysis using the Multiple Gamma ray detecting method (PGNAAMG) was estimated. As a result of this experiment, PGNAAMG can quantify to 0.05 ppm by the measurement for 10 minutes on the condition that the detector system have about 10% absolutely efficiency.

2P05: Utilization of pulsed neutron beams of JRR-3M: neutron spectrum measurement and discrete measurement of prompt and decay gamma rays

Matsue, H., Yonezawa, C. (JAERI, Tokai)

A neutron beam chopper has been installed in the JRR-3M prompt gamma-ray analysis system to generate pulsed neutron beams. Using the pulsed cold and thermal neutron beams, neutron spectrum measurement by a time of fight method and discrete measurement of prompt and decay gamma rays based on the lifetime difference of the excited nucleus have been studied.

2P06: Development and application of Cl-36 AMS using gas-filled magnet

Aze, T.,[a] Fujimura, M.,[a] Noguchi, M.,[a] Matsumura, H.,[b] Nagai, H.,[c] Matsuzaki, H.[d] ([a]Graduate School of Integrated Basic Sciences Nihon University, [b]High Energy Accelerator Research Organization, [c]College of Humanities and sciences Nihon University, [d]Research Center for Nuclear Science and technology, The University of Tokyo)

^{36}Cl-AMS system has been developed at MALT (Micro Analysis Laboratory, Tandem Accelerator), The University of Tokyo. We use a gas filled magnet(GFM) to separate ^{36}Cl from the interfering isobar ^{36}S. ^{36}S is suppressed by 3 orders by GFM. As a result, the sensitivity for ^{36}Cl/Cl ratio was about 10^{-13}. However this ratio is not sufficient for the measurements of natural samples. To overcome this situation, as well as to improve the reproducibility and the overall efficiency, we performed the Monte Carlo simulation of the behavior of ions n the GFM detector system.

2P07: Measurement of secondary cosmic-ray-induced ^{26}Al in silicate rock

Okuda, Y.[1], Sasaki, T.[1], Takamiya, K.[2], Oki, Y.[2], Shibata, S.[2], Matsuzaki, H. [3](Grad. School of Eng., Kyoto Univ.[1], Res. Reactor Inst., Kyoto Univ.[2], Res. Center of Nucl. Sci. Tech., Univ. of Tokyo[3])

In 1975, Hampel et al. reported the result of the measurement of secondary cosmic-ray-induced ^{26}Al (half life 0.716my) in silicate rock by low-level γ-γ coincidence spectrometry. In this work, we tried to measure ^{26}Al produced in silicate rock by accelerator mass spectrometry (AMS). Samples of chert were collected from Kutami, Yaotsu-Cho, Kamo-Gun, Gihu-Prefecture, Japan. Aluminum was chemically separated and purified from a few grams of chert. The ^{26}Al/^{27}Al ratio was measured by AMS using the tandem accelerator at Res. Center of Nucl. Sci. Tech., Univ. of Tokyo. The content of aluminum in chert was determined by neutron activation analysis using the research reactor (KUR) at Res. Reactor Inst., Kyoto Univ. As a preliminary result, secondary cosmic-ray-induced ^{26}Al in silicate rock was obtained to be 6-7 ($\times 10^{-4}$) dpm^{26}Al/1kg SiO_2.

2P08: Comparison of the amount of Au-198 induced by natural neutrons

Murata, Y.[a], Ahmed, M. Y.[a], Komura, K.[a] ([a]LLRL, Kanazawa Univ.)

To evaluate self-absorption of neutrons on gold activated by natural neutrons, some experiments relevant to gold thickness and the amount of ^{198}Au generated were performed. Many piled gold plates, size of which is 4 × 17.5 × 0.015, 4 × 15 × 0.012, 3 × 4 × 0.07, 4 × 5 × 0.02 and 3 × 4 × 0.035 cm, set into environment with shield and no shield, and on wall of reactor at UTR-KINKI. The gold plate samples were irradiated by natural neutrons for 3 weeks or by leakage neutron from reactor for 6 hours. ^{198}Au activities in the gold plates were measured by ultra low background γ-ray spectrometry at Ogoya Underground Laboratory. Although conditions of neutron irradiation were different, the grade of neutron self-absorption against gold thickness was almost the same among some experiments. However, compared with experiment performed before using the single gold plate, it was shown that ^{198}Au activities in piled gold plates were larger than those in single gold plates.

2P09: Radiocarbon dating of *Kohitsu-gire* fragments of the ancient manuscripts attributed to famous Japanese calligraphists

Oda, H.[a], Ikeda, K.[b], Masuda, T.[c], Nakamura, T.[a] ([a]Center for Chronological Res., Nagoya Univ., [b]Cyuo Univ., [c]Aichi Bunkyo Univ.)

Paper fragments of the old manuscripts written in *Heian-Kamakura* period are called *Kohitsu-gire*. Although they would provide important information for the study of classical literature and paleography, there are many counterfeits written a few centuries later among *Kohitsu-gire* attributed to the famous calligraphists. Therefore, to clear the written ages of 10 *Kohitsu-gire*, we measured their radiocarbon ages by the AMS method. Radiocarbon dating indicated that three samples are the counterfeits or copies written afterwards. Although two counterfeits had been believed to be genuine manuscripts by *Fujiwara no Teika* who is one of the most famous calligraphists, the result of radiocarbon dating excluded them from the materials for study on *Fujiwara no Teika*. On other *Kohitsu-gire* fragments, radiocarbon dating provided important information for paleography. In this study, it was shown that radiocarbon dating is a useful new method to determine

the written age of *Kohitsu-gire* fragments and to estimate the worth as the materials for the study of classical literature and paleography.

2P10: Determination of trace elements at sediment-water interface by INAA and ICP-MS

Kojima, S.,[a] Saito, T.,[b] Takada, J.,[c] Oda, H.,[d] Nakamura, T.,[d] Furukawa, M.,[e] Yokota, K.[f] ([a]Aichi Med. Univ., [b]Radioisotope Res. Center, Osaka Univ., [c]Res. Reactor Inst., Kyoto Univ., [d]Center for Chronol. Res., Nagoya Univ., [e]Yokkaichi Univ., [f]Lake Biwa Res. Inst.)

Trace elements in sediment and pore water were measured by instrumental neutron activation analysis (INAA) and inductively coupled plasma mass spectrometry (ICP-MS) in this study. Sediment reference material samples (JLk-1, the Geological Survey of Japan) irradiated with reactor neutrons were dissolved by conventional hot plate digestion method with an acid mixture, and complete decomposition was confirmed by detection of γ-rays from the filtrate and the residue. Sediment core samples were taken on board with a gravity corer in Lake Biwa. Pore water samples were separated from the sediments by centrifugation. Thirty-two elements in the sediments could be determined by ICP-MS, including the eight elements of Mg, P, Ti, V, Ni, Cu, Sr and Pb, which were difficult to be determined by INAA. Measurement of trace elements in the pore water samples is undergoing.

2P11: Effect of presence of pine trees on the variation in elemental composition of airborne dust

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

For the research on environmental impact caused by nuclear activities, we are developing monitoring technique to measure radioactivity, isotopic and elemental composition in airborne dust, which contains nuclear materials and radioactive elements released from nuclear facilities. We focus on its elimination process such as wash-out by rainfall and adsorption to plants in the processes which affect the amounts and composition of the airborne dust. Recently at JAERI-Tokai, a number of pine trees to arrest sand shifting was cut down. This gave us a good opportunity to examine the effect of presence of the trees on the variation in elemental composition of airborne dust, therefore, we collected dust samples with a high-volume sampler before and after felling the trees, and analyzed them by means of instrumental neutron activation analysis. No change was observed in the amount of the dust collected before and after the trees felling nearby the sampling site, while the Sc/Na–Cl/Na plots showed that the sea-aerosol could be easily transported to the sampling site after felling the trees. At the presentation, the variation in concentration of other elements will be reported.

2P12: Activation analysis of air particulate matters

Oura, Y.[a], Nakamatsu, H.[a], Nagahata, T.[a], Otoshi, T.[b], Ebihara, M[a] ([a]Tokyo Metropolitan Univ., [b]Tohoku Univ. of Comm. Serv. Sci.)

Chemical compositions of air particulate matters (corresponding to $PM_{10-2.5}$ and $PM_{2.5}$) collected at Hachioji, Tokyo, and Sakata, Yamagata prefecture, were determined by prompt gamma-ray analysis (PGA) and k_0 standardized instrumental neutron activation analysis. Almost reference elements for artificial origin particulates, were enriched in fine particulates fractions ($PM_{2.5}$) at Sakata. At Hachioji, a few reference elements for artificial origin particulates were enriched in fine particles fraction like at Sakata, but many reference elements for artificial origin particulates were distributed approximately equally in both fine and coarse particulates. As one possible its cause, Hachioji area was estimated to have more kinds of origin of artificial particulates than Sakata area. Boron concentrations determined by PGA at Hachioji and at Sakata were almost same level and it was found that B was enriched in fine particles. This fact suggested that B in air particulate matters were mainly originated from artificial particulates.

2P13: Fluctuation of several element concentrations in cherry blossom during blooming

Takada, J., Kawamoto, K. (Res. Reactor Inst. Kyoto Univ.)

We have studied the change of element concentration in the petal of cherry blossom during blooming. The flower of cherry is a favorable material to examine the change of the element distribution with the growth of the flower. The samples were collected at a constant time interval between bud formation and full blossom. Petal materials were irradiated in Pneumatic tube of Kyoto University Reactor, and the concentrations of several elements were determined of neutron activation analysis. The relative concentration in petals was increased in alkaline and rare earth during blooming. On the other hand, elements with flower blooming, while that of Ca, Ba and halogen elements tended to decrease. These trace elements seemed to be an important role in the blooming process.

2P14: Concentration Variations of ^{210}Pb, ^{7}Be, ^{40}K and some elements in plants leaf samples

Efrizal, Sugihara, S., Osaki S.,[1] Maeda Y. (Department of Chemistry and Physics of Condensed Matter, Graduate School of Sciences, Kyushu University, [1]Radioisotope Center, Kyushu University)

The activities of the natural radionuclides ^{7}Be, ^{210}Pb and ^{40}K in leaf samples of eight plants were measured. These three natural radionuclides are useful to investigate the distribution of the radionuclides in environment. The leaf samples were monthly collected in Mt. Sefuri (Fukuoka Prefecture) and the sample had been ashed at 550°C. Radioactivities of ^{7}Be, ^{210}Pb, and ^{40}K were measured with a gamma-ray spectrometer using a Ge-semi-conductor detector, and some elements were determined by a neutron activation analysis. Seasonal change of ^{7}Be and ^{210}Pb accumulations in the samples was observed and each sample shows characteristic variation pattern: the activities were generally increased from August to April/May. Especially ^{40}K in sample *Petasites japonicus* and *Miscanthus sinensis* shows clear pattern where decrease from May through next March. On the other hand, the concentrations of elements Fe, Co, Cr and Mn are constant. Correlation factor (R) of ^{210}Pb with ^{7}Be indicated both of radionuclides have same source from deposition of aerosol.

2P15: Absorption behaviors of technetium and rhenium through plant roots

Tagami, K., Uchida, S. (Natl. Inst. Radiol. Sci.)

The absorption behaviors of technetium (Tc) and rhenium (Re) through plant roots were studied using nutrient solution culture. Radish samples, grown in nutrition culture for 20-30 days in a green house, were transferred into plastic vessels containing nutrient solutions contaminated with multitracer solution including Tc-95m and Re-183. The plant samples were grown individually for 1-7 days in a laboratory condition. The activities in nutrient solutions and oven-dried plant parts (roots, fleshy roots and leaves) were measured with Ge detecting systems. The concentrations of Tc-95m and Re-183 in the nutrient solutions after harvesting the plants were almost the same with those in the initial solution. Possibly, the radionuclides were taken up with water through plant roots. The distributions of Tc and Re in the plants showed no differences, thus, soluble Tc and Re absorption behaviors by plant samples were the same. It was suggested that Re could be used as a geochemical tracer of Tc in the soil environment.

2P16: Detection of uranium and thorium disequilibrium in phosphate fertilizer

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

We reported last year a large radioactive disequilibrium between U-234 and U-238 in a commercial phosphate fertilizer. The sample fertilizer was leached last year in a column successively by using

water, 0.1 M HCl, and 8 M HCl, and then the residue was digested and leached with a mixture of hydrofluoric acid, nitric acid and perchloric acid in a pressurized PTFE container. The ratio of U-234 / U-238 radioactivities amounted to 7.6+/-0.9 in the final fraction of successive leaching steps. This year we introduced the method of batch leaching with water, 0.1 M HCl, and 8 M HCl in order to reduce processing time and to get additional information upon the radioactivity of Th-230. But little disequilibrium was observed in the fertilizer with the same lot number in even uranium isotopes till now. Possible explanations for this result are: 1) heterogeneity of the sample fertilizer, or 2) different behavior of isotopes to the chemical processes (column / batch leaching method). It is hard to understand the second one.

2P17: Analysis of the Nagasaki Omura bay bottom sediment using environmental radioactivity

Fujiyasu, N.,[1] Yoshikawa, I.,[2] Takatsuji, T.,[2] Nakamura, M.[1] ([1]Grad. Sch. Env. Sci., Nagasaki Univ., [2]Fac. Env. Sci., Nagasaki Univ.)

Omura bay in Nagasaki Pref., Japan is restrictedly isolated from the outer sea. The content of Pb-210 in the bottom sediment separated horizontally at 1cm interval of the Omura bay was measured using an n-type high-purity germanium detector with very thin window (Gamma-X detector). From the measurement, the average sedimentation rate at the bottom was estimated to be 110 $mg/cm^2 \cdot y$, and higher estimations were derived at the northwest part (the former "Holland Village") and the southwest part (near the mouth of the Shishigawa River) of the bay. The Pb-210 flux at the bottom was estimated in the range 90 ~ 325 $Bq/m^2 \cdot y$, and the northwest part showed the higher flux as same as the sedimentation rate. Recently, we analyzing the heavy metals element in sediment. The result may help to reveal the chronological change of the environment surrounding the bay.

2P18: The behavior of ^{210}Pb and ^{7}Be in the atmosphere in Nagasaki

Nakamura, M.,[a] Yoshikawa, I.[b] Takatsuji, T.[b] Fujiyasu, N.[a] ([a]Grad. Sch. Env. Sci., Nagasaki-Univ., [b]Fac. Env. Sci., Nagasaki-Univ.)

Nagasaki is located in the western end of Japan, and first receives aerosol factors in Japan from the Eurasian Continent such as Asian dusts and directly almost with no mixing of the domestic factors. Since this geographically situation of Nagasaki area, we have measured the aerosol size and the concentration of ^{210}Pb and ^{7}Be adhering to aerosol. Seasonal variation of these data was analyzed combined with some weather data, the rainfall and the wind direction. We present and discuss some result from the analysis.

2P19: Study on environmental change recorded in the sediment of Nishiyama reservoir at Nagasaki

Saito, Y.[1], Esaka, F.[1], Yasuda, K.[1], Magara, M[1]., Miyamoto, Y.[1], Sakurai, S.[1], Usuda, S.[1], Yamazaki, H.[2], Yoshikawa, S.[3], Nagaoka, S.[4] ([1]JAERI, [2]Kinki Univ., [3]Osaka City Univ., [4]Nagasaki Univ.)

More than 50 years have passed since the atomic bomb attack in Nagasaki and its traces has been fading. Yoshikawa et al. reported the possibility of determination of Black rain recorded in the sediment (A core) of Nishiyama reservoir at Nagasaki. To make a further search for the traces of the atomic bomb, another sediment (B core) excavated at the same time was analyzed in this study. We determined radioactivities and elemental concentrations as well as their chemical forms. It was observed in depth distribution that B core had two peaks of Cu and Pb concentrations, which were similar to the profile of ^{137}Cs. This result was different from that reported previously by Yoshikawa et al. The 225 cm-depth sample, which had a peak of Cu concentration, didn't contain particles of Pu but contained those of Cu and its alloys. It was considered that these particles were artificial and were indirect evidence of the trace of the explosion.

2P20: Retrospective dosimetry of quartz and feldspar extracts from atomic bomb-suffered samples using RTL- and BTL-measurements

Nomura, S.,[a] Hashimoto, T.,[b] ([a]Graduate School of Science and Technology, Niigata University., [b]Faculty of Science, Niigata University)

Thermoluminescence technique has been used for dating archaeological samples. It will be also useful for retrospective dosimetry when the artificial radiation doses are separable from naturally accumulated doses. The quartz and feldspar grains were extracted from roof tiles and porcelains, which were collected from underground in the Hiroshima Peace Memorial Park. The accumulated doses from such extracts were evaluated by TL measurements using SAR protocol. In the case of quartz extracts, simultaneous RTL- and BTL-measurements were initially applied in this experiment and the retrospective dose was estimated to be about 50Gy in good agreement with each other. In contrast, RTL measurement from feldspars resulted in the less dose evaluation about 30Gy. From the porcelain disc prepared for RTL measurements, the estimated dose corresponded to be about 10% of doses from the quartz grains in roof tiles. The radiation-shielding effect was considered to cause the difference between them. In comparison with the estimation of the naturally accumulated doses, the evaluated γ-dose should be entirely derived from the atomic bomb.

2P21: Characterization and geochemical behaviors of uranium series nuclides in sedimentary rocks near the boundary area of Niigata and Yamagata Prefectures

Kanai, Y., Kamioka, H., Nakajima, T., Tsukamoto, H. (Geological Survey of Japan, AIST)

Uranium series nuclides were studied in sedimentary rocks from Nakamaruke district (Niigata Pref.) and Kanamaru district (Yamagata Pref.). At Nakamaruke, uranium was correlated with the phosphate and enriched in the bentonite layer below the phosphatic layer. A large disequilibrium ((U-234/U-238<1, Th-230/U-234>1) was observed in the phosphatic layer. The samples were treated with nitric acid and separated as acid-soluble and insoluble fractions. Most of uranium was present in the acid soluble components. Uranium in the acid insoluble fraction was enriched with U-234, probably by the alpha recoil effect. At Kanamaru on the other hand, uranium had no correlation with phosphate. There were two U-enriched layers in arkose sand layers (Kamagui Group), and one of them was in radioactive disequilibrium (Th-230/U-234>1). Some samples showed the radioactive disequilibrium and it is inferred that uranium has moved within 0.3 million years.

2P22: Uranium in mine drainage from the disused Ogoya Mine in Ishikawa Pref., Japan - Unusually high $^{234}U/^{238}U$ activity ratios -

Yamamoto, M.,[1] Sakaguchi, A.,[1] Kofuji, H.,[2] Oda, H.,[3] Nakamura, T.[3] ([1]Kanazawa Univ., LLRL, [2]JMSF Mutsu Marine Inst., [3]Nagoya Univ., DMRC)

From the viewpoint of the radioactive disequilibrium in U-series, U concentrations and their $^{234}U/^{238}U$ activity ratios have been measured for surface and underground waters around the disused Ogoya Mine in Ishikawa Pref., Japan. The concentrations of ^{238}U and their activity ratios of $^{234}U/^{238}U$ were found in the wide range of 0.004-0.8 mBq/L and from ca.2 to as high as 16, respectively These values were the highest in the drainage from the mine, which is strong acidic wastewater (pH 3.5-4.0). Generally, such a high $^{234}U/^{238}U$ activity ratio has been explained by the preferential leaching of recoil-derived ^{234}U from rock materials into the ground water under reduced condition near neutral water. Here, it is scientifically interesting to clarify the reason why the $^{34}U/^{238}U$ activity ratio is unusually high in spite of the high ^{238}U

concentrations (0.5-0.8 mBq/L) under the acidic condition. Detailed discussion will be given in connection with the formation of acidic wastewater and the residence time of its water estimated by carbon isotopic measurements (AMS).

2P23: Correlation between plutonium concentration in soil and in humic substances from Japan

Fujikawa, Y.[a], Ikeda, E[b], Sugahara, M.[b] ([a]Res. Reactor Institute, Kyoto Univ., [b]Osaka Sangyo Univ.)

We measured the concentration of global fallout plutonium (Pu) in soil and soil humic substances (HS) extracted and purified from organic horizons of brown forest soil from Nishiyama, Nagasaki, Japan and Humic Andosols in Yatsuka, Okayama, Japan. Pu was associated with the HS as a result of *in-situ* reaction in surface soil, and only Pu that was strongly associated with HS was analyzed because the HS was thoroughly purified to remove inorganic impurity by repeated treatment with acid prior to Pu measurement. Assay of Pu in soil and in the HS showed that while the Pu concentration in soil varied depending on the location, the Pu content of humic acid (HA) was linearly correlated with Pu concentration in bulk soil (correlation coefficient $r^2 > 0.9$). Such significant correlation, on the other hand, was not found for the other elements (Cu and U).

2P24: Determination of uranium and thorium in iron compounds by alpha spectrometry

Takano, M.,[a] Okada, Y.,[a] Hirai, S.,[a] Mitsugashira, T.,[b] Hara, M.[b] ([a]Fac. of Eng., Musashi Inst. of Tech., [b]Inst. for Material Res., Tohoku Univ)

Uranium and thorium nuclides in JSS red iron ore reference material, JSS 805-1 and JSS 803-4, were determined by applying the coprecipitation method with SmF_3 (Sm method). The analytical samples were decomposed by a mixed acid solution of hydrochloric acid and hydrofluoric acid. Uranium and thorium were coprecipitated with neodymium fluoride to remove matrix iron. Then, uranium and thorium were purified and isolated separately by an anion-exchange chromatography. The uranium and thorium were finally collected in SmF_3 precipitates and the precipitates were mounted as alpha-spectrometric samples. The results of alpha-spectrometry were compared with the results of INAA and we confirmed that the chemical recovery yield was quantitative. It was found that ^{232}Th and ^{228}Th were in radioactive equilibrium in red iron ore samples but ^{230}Th was not in radioactive equilibrium with its precursor ^{238}U and ^{234}U.

2P25: On the emission of neutrons from uranium hexafluoride

Furukawa, M.,[a] Komura, K.,[b] Imanaka, T.[c] ([a]Yokkaichi Univ., [b]LLRL, Kanazawa Univ., [c]Res. Reactor Inst., Kyoto Univ.)

We observed that excess neutrons were emitted from the building where low-enriched uranium was stored as fluoride: The emission was studied by measuring millibecquerel level of ^{198}Au induced in gold metal which had been placed outside of the building. The phenomenon was examined taking spontaneous fission of ^{238}U and ^{19}F(α, n) reaction into account. Nehurukawa utron emission from 1kg of ^{238}U was calculated to be 15 /s from spontaneous fission: Whereas that from 1kg of low-enriched uranium (^{235}U, 4.0%) was estimated to be 130/s by ^{19}F (α, n) reaction. We concluded that the contribution of the nuclear reaction was much larger than that of spontaneous fission. However, neutron emission from low-enriched uranium stored as oxide was far less than the emission from that stored as fluoride.

2P26: Learning of and participation in radiations based on awareness and voluntaryism

Murabayashi, K.[a], Aratani, M.[b] ([a]Science Volunteer R, [b]Inst. for Environmental Sciences)

Mutsu-Ogawara Development and construction of nuclear industry facilities in the Penin. Shimokita (the nuclear power peninsula) started before about 30 years are still in progress, and are discussed positively from viewpoint of how to make the best of them for activation of the regional society. After the JCO neutron criticality accident, awakening as residents of the most important site in the energy policy of Japan changed their consciousness from "people to be informed" to "people to try to know". On this May the Pacific side of the Penin. Shimokita, Aomori has been authorized by the government as a specified area for creation of environmental and energy industry, originally the proposal of which had been made by the residents. Several activities will be reported. 1) Penin. Shimokita Activation Society. 2) Rokkasho Reading Circle. 3) Non-Profit Organization Environmental Guardian of Gaia. 4) Aoimori Science BBL (Brown Bag Lunch).

2P27: Recent status of research activities for development of CTBT-related technologies at JAERI

Shinohara, N., Hirota, N., Houkida, T., Inoue, Y., Kumata, M., Kurosawa, Y., Miyamoto, Y., Nakahara, Y., Oda, T., Uchikoshi, T., Yamamoto, Y. (JAERI)

Research activities of the R & D Group for Non-Proliferation Technology related to the Comprehensive Nuclear-Test-Ban Treaty (CTBT) verification regime are presented in the Symposium. The main subjects of this presentation are (i) an overview of the CTBT verification regime, (ii) preparation of the National Data Center for radionuclide data, (iii) construction and operation of the radionuclide monitoring stations at Takasaki and Okinawa and the certified rarionuclide laboratory at Tokai in Japan. We have participated in an intercomparison test internationally organized for the certified laboratories and the test results are given here. Scientific application of the CTBT-related technologies to environmental researches is also explained.

2P28: An examination of the alpha sources prepared by samarium coprecipitation method under a scanning electron microscope

Kikunaga, H.,[1] Mitsugashira, T.,[2] Hara, M.,[2] Kasamatsu, Y.,[3] Shinohara, A.[3] Takamiya, K.,[4] Ohtsuki, T.,[5] Yuki, H.,[5] Kinoshita, N.,[1] Yokoyama, A.,[6] Nakanishi, T.,[6] ([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]The Oarai-branch, Inst. Materials Res., Tohoku Univ., [3]Grad. School Sci., Osaka Univ., [4]Res. Reactor Inst., Kyoto Univ., [5]Lab. Nucl. Sci., Grad. School Sci., Tohoku Univ. [6]Faculty Sci., Kanazawa Univ.,)

Samarium coprecipitation method is simple and easy for preparation of the alpha source giving a high resolution of energy spectra with a semiconductor detector. The method is utilized in measurement of the alpha activity from actinide elements in our project. The energy resolution depends on the preparation conditions such as selection and quantity of coprecipitation reagent. In this study, we attempted to optimize the condition of alpha source preparation with the use of a scanning electron microscope, which enables one to observe a microscopic view of the precipitates corresponding to the source qualified for highly resolved alpha spectrometry.

2P29: Variation of radon concentration in the groundwater flowing out from small mountainside area

Horiuchi K.[a], Kobayashi M.[b] ([a]Otsuma Women's Univ., [b]Dept. Natural Sci., Osaka Kyoiku Univ.)

From June in 1998, we have studied continuous radon concentration in groundwater from the 3 different types of small mountainside area (Basin I, II, III) which including a landslide and an artificial change area. At same time we have analyzed some kinds of chemical elements such as Na^+, K^+, Mg^{2+}, Ca^{2+}, HCO_3^-, Cl-, NO_3^-, SiO_2 and others. At the Basin I (natural area), we did not

measure radon concentration, because of the shortage of sample water. Continuous measuring of Radon concentration and the water temperature in the groundwater of Basin II (artificial change area) shows clear seasonal variation. For draining groundwater from the Basin III (landslide area), the horizontal 13 holes, from N1 to N13, radiating in all directions were bored through the river side of the Basin III. But only from the N1, N6, N7, N8, N9, N13 holes, groundwater flow out regularly. According to the season, amount of radon and temperature of N1 and N6 holes water have changed, but the other holes water have not shown clear variation. In conclusion, we have reasoned that the almost groundwater in the Basin II would depend on the surface water and in the Basin III, N1 and N6 holes groundwater would mainly depend on the shallow free water and the other holes one on the deep confined water.

2P30: Continuous observation of atmospheric concentrations of ^{212}Pb and ^{214}Pb in Kawasaki, Japan

Kukita, K., Koike, Y., Nakamura, T., Sato, J. (School of Sci. and Technol., Meiji Univ.)

Atmospheric concentrations of ^{212}Pb and ^{214}Pb were observed with aerosol samples in Kawasaki. In order to discuss temporal behavior of radon- and thoron-progeny, diurnal variations of the radioactive lead isotopes were determined by low energy photon spectrometer (LEPS). In Kawasaki, diurnal concentration patterns of ^{212}Pb were similar to those of ^{214}Pb. It is noted that the concentration of ^{212}Pb is correlated with that of ^{214}Pb. The atmospheric concentrations of ^{212}Pb and ^{214}Pb were significantly affected by meteorological parameters, especially wind velocity.

2P31: Secular change in atmospheric $^{14}CO_2$ in the south area of Osaka Prefecture II

Shibata, S. Kawano, E. (Radiation Res. Center, Osaka Prefecture University)

We continued measuring atmospheric $^{14}CO_2$ by NaOH method at Radiation Res.Center of Osaka Pref.Univ. (Sakai city) for 16 years from 1985 to compare with ^{14}C of rice grains which harvested at the campus fields. The measurement of atmospheric $^{14}CO_2$ at Izumi city adjacent to Sakai city was also continued for five years from 1992.There are two graphs. Fig.1 shows the changes of atmosphere $^{14}CO_2$ at Sakai and of ^{14}C of the rice grains. It is clearly shown that atmosphere $^{14}CO_2$ in Sakai is high in summer and low in winter. Fig.2 shows atmospheric $^{14}CO_2$ change at Izumi. There is a slight difference in atmospheric $^{14}CO_2$ change in both cities.

2P32: Significant features of gamma-ray dose increase associated with winter thunderstorms

Yamazaki, K.,[a] Ohtaka, T.,[b] Fujimaki, H.,[a] Tonouchi, S.,[a] Tanabe, H.,[a] Hashimoto, T.[c] ([a]Niigata Pref. Inst. Environ. Rad. Mon., [b]Inst. Public Health & Environ. Sci., [c]Niigata Univ.)

Around the Kashiwazaki Kariwa nuclear power station, 58 particular radiation-induced dose-rate peaks associated with winter thunderstorms have been observed in the time variation of environmental gamma-ray dose rates measured by IC (ionization chamber) detectors during the past 5 years. Ten of them were accompanied with the dose-rate increase detected by NaI(Tl) scintillation detectors. Therefore, the both detectors seemed to be exposed to some instantaneous radiation at the moments. The following features have been extracted from these 10 cases: (1) The ratios of the dose rate increment of NaI to that of IC are much smaller than unity. (2) The pulse-height distribution measured by NaI detector was counted up to a high-energy range, unlike that of other cases. However, there is not an adequate correlation between the increment of IC and that of count rates beyond 3MeV. (3) The observed position of dose-rate increase seems to move along with the active region of thunderclouds. According to these results, it is supported that Bremsstrahlung X-rays generated at high altitude has been attributed to the radiation source.

2P33: Transfer of atmospheric water vapor to dead leaf - experiments using deuterium

Momoshima, N.,[a] Matsushita, R.,[a] Nagao, Y.,[a] Ichimasa, Y.,[b] Ichimasa, M.[b] ([a]Kumamoto Univ., [b]Ibaraki Univ.)

Dead leaves of 4 plant species were exposed to deuteriated (D) water vapor at daytime and nighttime to examine water vapor transfer from atmosphere to dead leaf. The experiment is carried out in relation to environmental safety for nuclear fusion facility that will use a large amount of tritium as fuel. The D concentrations in the dead leaf reached 4 to 87 % of those in the air after 1 or 2 h exposure, the daytime exposure showed drying of leaf and lower D concentrations compared to the nighttime exposure. The species having larger number of stoma showed higher D concentrations both daytime and nighttime exposures, suggesting number of the stoma controlling the D uptake. The open and close of stoma, which occur in relation to water content of stoma cell, is also important factor for D uptake even on dead leaf as already known on live leaf. Reduced uptake of D on the daytime exposure is attributed to close of stoma due to lower water content in leaf.

2P34: Development of a tritium monitor in the Air

Sakuma, Y.[a], Koganezawa, T.[b], Iida, T.[b], Ogata, Y.[c], Aoyma, T.[c], Torikai, Y.[d], Tanaka, M.[e], Ohta, M.[f], Takami, M.[f]. ([a]NIFS, [b]Graduate School of Eng., Nagoya Univ., [c]School of Health Sciences, Nagoya Univ., [d]Hydrogen Isotope Res. Center, Toyama Univ., [e]Japan Air-conditioning Service Co.&Ltd., [f]Fac. of Eng., Niigata Univ.)

We are developing a tritium monitor in the air which is able to measure tritiated water vapor (HTO), tritiated hydrogen (HT) and tritiated methane (CH_3T) separately. The concentration of hydrogen and methane in the air are ~0.5ppm and ~1.5ppm respectively. However, these specific activity are much larger than the specific activity of water vapor. Then, we are trying to collect hydrogen and methane from the air, and developing a sensitive proportional counter to measure them.

2P35: Protecting effect of natural antioxidants against lipid peroxidation induced by radiation. -Effect of the location of initiating radicals-

Tsuchiya, H.[1], Taguchi, H.[1], Saitoh, T.[3], Warashina, T.[3], Yoshioka, H.[2], Yoshioka, H.[1] ([1]Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ., [2]Res. Reactor Inst., Kyoto Univ., [3]Inst. For Environmental Sci., Univ. of Shizuoka)

Recently, attention has been focused on the natural antioxidants being able to scavenge reactive oxygen species which induce aging, cancer and the other diseases. In the previous paper, inhibiting effect of tea catechins on radiation-induced lipid-peroxidation was investigated. It was considered that the hydroxyl radical (HO·) formed by gamma-irradiation near the surface of lipid membrane acts as a initiator of peroxidation. However, various radicals generated in the membrane, on the surface and in the outer water phase can be the initiator and the inhibiting activity changes depending on the mutual positioning of the initiator and the antioxidant. In this experiment, AMVN which produces a radical in the lipid membrane was used and the results were compared with those of gamma-irradiation. A lipophillic antioxidant α-tochopherol worked effectively for scavenging radicals locating in the membrane.

2P36: Short-lived radicals of DNA bases formed by the reaction with the hydroxyl radical (II)

Taguchi, H.,[1] Tsuchiya, H.,[1] Yoshioka, H.,[2] Yoshioka, H.[1] (Faculty of Science, Shizuoka University[1] and Institute for Environmental Sciences, University of Shizuoka[2])

The mechanism of the DNA damage by the hydroxyl radical (·OH) produced with the radiation on water was investigated using the rapid-flow ESR method. In this experiment, (Ti^{3+} + H_2O_2) system was applied for ·OH generation instead of radiation and the

radicals of DNA bases, thymine, cytosine, adenine and guanine, formed by the reaction with the ·OH were examined. The radicals of pyrimidine bases, namely thymine and cytosine, showed a double quartet and a triplet spectra, respectively, showing that ·OH was attached to the C=C double bond and the unpaired electron located on a carbon atom. In the case of the other purine bases, a weak, broad signal was observed, suggesting that dehydrogenation by ·OH occurred and, as a result, planar character of the molecule was maintained resulting in the dislocation of the unpaired electron.

2P37: Formation mechanisms of $TcO_2 \cdot nH_2O$ nanoparticles by bremsstrahlung irradiation of TcO_4^-

[a]Sekine, T., [a]Suzuki, T., [a]Takayama, T., [a]Kudo, H., [b]Lin, M., [b]Katsumura, Y. ([a]Tohoku Univ., [b]Univ. Tokyo)

$TcO_2 \cdot nH_2O$ nanoparticles were produced by bremsstrahlung irradiation of aqueous TcO_4^- solutions and its formation mechanism was investigated. $TcO_2 \cdot nH_2O$ nanoparticles were formed in the target solutions at pH > 3, while a soluble Tc(IV) species ($Tc^{IV}O_2$ polymers) having an absorption peak at 320 nm was produced in acidic solutions (pH < 3). $TcO_2 \cdot nH_2O$ nanoparticles were also formed when pH of the irradiated solution containing the polymer was increased. The result suggests that the polymer should be a precursor of $TcO_2 \cdot nH_2O$ nanoparticles. Yields of the Tc(IV) species produced by irradiation were simulated with a FACSIMILE program based on reduction processes involving successive disproportionation reactions of Tc(VI) and Tc(V) followed by a bimolecular reaction of a TcO_4^- ion and a H radical. The experimental results were well reproduced by the simulation with a rate constant of $5 \times 10^7\ M^{-1}s^{-1}$ for the reaction of TcO_4^- with H.

2P38: Synthesis of ^{227}Th-EDTMP and measurement of its biodistribution

Washiyama, K.[a], Kinuya, S.[a], Mitsugashira, T.[b], Amano, R.[a] ([a]Fac. Med., Kanazawa Univ., Institute for Material Research, Tohoku Univ.)

Thorium-227 ($T_{1/2}$=18.72 day) is an alpha emitter and its daughter is a ^{223}Ra ($T_{1/2}$=11.435 day), which is well known nuclide for therapy of bone metastases. When ^{223}Ra has accumulated in bone, almost daughter nuclides are retained in bone and emit multiple alpha and beta radiations. In this study, we synthesized ^{227}Th-EDTMP and evaluated the biodistribution of it for the efficacy of radiopharmaceutical for bone metastases. ICR male, 7-week-old, mice were administered 0.1mL of ^{227}Th-EDTMP. After appropriate time intervals, the liver, kidney, spleen, lung, muscle, femur, parietal bone and blood are excised and subjected to gamma-ray spectrometry. As the results, almost ^{227}Th were accumulated in bone and were retained during 14days. Uptake of other tissue was few and gradually decreased till 14 days. These results showed that ^{227}Th-EDTMP is a good candidate for therapy of bone metastases.

2P39: An application of bismuth isotopes for nuclear medicine

Washiyama, K.[a], Kanayama, Y.[a], Kinuya, S.[a], Haba, H.[b], Enomoto, S.[b], Amano, R.[a] ([a]Fac. Med., Kanazawa Univ., [b]RIKEN)

Bismuth isotopes belong to decay-chain nuclides are of considerable interest for tumor-therapy in nuclear medicine due to their alpha-decay mode. However it is difficult to calculate the radiation dose experimentally due to their short half-life. Bismuth-206 has a long half-life and can be detected by its characteristic gamma-rays. Therefore, it has been thought as a suitable isotope for alternative tracer for short-life Bi isotopes. In this study, we made a ^{206}Bi tracer by ^{nat}Pb (p, xn) ^{206}Bi reaction with proton accelerator and compared the biodistribution of ^{206}Bi in mice with ^{213}Bi in order to evaluate the radiation dose of Bi isotopes belong to decay-chain nuclides. Mice were administered 0.1mL of ^{206}Bi solution intravenously. After appropriate time intervals, the liver, kidney, spleen, stomach, femur and blood are excised and subjected to gamma-ray spectrometry. As the results, almost ^{206}Bi were accumulated in kidney and other tissue had few uptakes. These trends were same to the ^{213}Bi injection experiment. ^{206}Bi may be an alternative tracer for Bi isotopes belong to decay-chain nuclides.

2P40: Olfactory transport of alkaline metal ions by intranasal administration (1) —Direct brain uptake of $^{86}Rb^+$ and $^{137}Cs^+$—

Kanayama, Y.,[1] Yanagida,[1] T., Amano,[1] R., Enomoto, S.[2] ([1]Kanazawa Univ., [2]RIKEN)

Olfactory transport enables variable solutes to deliver from the nasal cavity directly to the brain. Our previous works, multitracer screening on inorganic ion delivery manner, revealed that intranasally administered ^{83}Rb was directly accumulated in the brain. Subsequently, ^{86}Rb intranasal behavior demonstrated that the Rb delivery was due to the olfactory transport. It is well known that Rb^+ is an alkaline metal ion and mimics K^+ biobehavior, so other alkaline metal ions could move through the olfactory route into the brain. In the present work, unilateral intranasal administration of $^{137}Cs^+$ physiological saline caused ipsilateral accumulation not only in the nasal mucosa but also the olfactory bulb. Furthermore, the ^{137}Cs, accumulated in the olfactory bulb unilaterally, distributed in the other part of the brain ipsilaterally. These results clarified that the intranasally administered $^{137}Cs^+$ was directly delivered to the brain, and showed the differences between the olfactory transport of Rb^+ and Cs^+. The olfactory transport of K-mimic ions could be an important knowledge for the rhinological or neurobiological studies of olfactory nerve systems.

2P41: On-line isothermal gas chromatographic behavior of Rf (Z=104) an its homologs (Zr, Hf)

Kaneko, T.[a], Tsukada, K.[a], Akiyama, K.[a], Asai, M.[a], Ishizu, H.[b], Ono, S.[b], Hirai, T.[b], Goto, S.[b], Haba, H.[c], Nishinaka, I.[a], Toyoshima, A.[d], Ichikawa, S.[a], Nagame, Y.[a], Kudo, H.[b] ([a]Tandem Accelerator Group, JAERI, [b]Advanced Sci. Res. Center, JAERI, Facl. Of Sci. [b]Niigata Univ., [c]Cyclotron Center, RIKEN, [d]Osaka Univ.)

The behavior of Zr and Hf in the gas system Cl_2/CCl_4 on SiO2 surface was investigated using the methods of online isothermal gas chromatography. The adsorption data of their chlorides were evaluated. It was found that Zr and Hf chlorides behaved similarly in this system. Therefore, the adsorption behavior of chlorides of these elements for quartz surface is not differing each other. That is reasonable for expectation from sublimation enthalpy in macro scale. And in the experimental condition, the behavior of Rf chlorides was investigated. It was found that the volatility of Rf chloride resemble with Zr and Hf.

2P42: JAERI-KEK joint RNB project

Kaneko, T., Matsuda, M., Osa, A., Ichikawa, S., Yoshida, T. (JAERI)

In accordance with the Japan Atomic Energy Research Institute (JAERI) – High Energy Accelerator Research Organization (KEK) joint radioactive nuclear beam (RNB) project, an ISOL-based radioactive nuclear beam facility is under construction to provide RNBs having energy up to 1.1 MeV/u at JAERI tandem accelerator. Uranium carbide target will be used for the production of neutron-rich nuclides as RNBs, we are developing a cavity type thermal ion source and FEBIAD type ion source for JAERI-ISOL. For using the new ion sources, an ion source housing and an acceleration chamber were changed completely.

2P43: The effect of the hydrogen halides on the yield of Re-186-labelled radiopharmaceuticals

Nogawa, N., Okuda, A., Makide, Y., Morikawa, N., Sato, A.[a] (Radioisotope Center, The University of Tokyo, [a]JAERI)

Re-186 (β-ray: 1.07 MeV, $T_{1/2}$: 3.8 d) is expected to be effective for pain palliation of skeletal metastases and cancer therapy.

Synthesis condition of Re-186-labelled radiopharmaceuticals requires a larger quantity of $SnCl_2$, lower pH of solution, higher temperature and longer reaction time in comparison with that of Tc-99m-labelled radiopharmaceuticals. The effect of hydrogen halides on the reaction yield was examined in order to attain milder and more convenient conditions. Reaction between $^{186}ReO_4^-$ solution produced by JAERI and DTPA, DMSA, or D-Pen was studied. Diluted HF, HCl, HBr or HI was used as a solvent. With the concentration of Cl^- and Br^- in the solvent, the yield increased. The reactivity of Re seems to be accelerated by the coordination of these halide ions instead of OH^- in Re ions.

2P44: Labeling of bifunctional chelating agent, MAG3, with ^{188}Re for radiotherapy

Hashimoto, K.[a], Wan Kamal, W.H.B.B.[b], Matsuoka, H.[a] ([a]Japan Atomic Energy Res. Inst., [b]Malaysian Inst. for Nucl. Tech. Res.)

The radioisotopes of rhenium (^{186}Re and ^{188}Re) are attractive radionuclides for radiotherapy because of their energetic beta particles and gamma rays suitable for imaging. Mercapto-acetyltriglycine, MAG3 is a useful bifunctional ligand in labeling monoclonal antibodies with metallic radionuclides. In this study, the labeling of MAG3 with carrier-free ^{188}Re from a ^{188}W/^{188}Re generator was investigated in detail. The ^{188}Re-MAG3 complex was synthesized by the direct labeling method and by the indirect labeling method using a transfer ligand (citrate or gluconate). The dependence of the labeling yield upon the reaction conditions such as the concentrations of the reducing agent, MAG3 and the transfer ligand, pH, temperature, reaction time and the addition of a carrier was examined. Under the optimum conditions, the labeling yield of ^{188}Re-MAG3 synthesized by the all method was over 90%. The differences of the optimum conditions such as pH and reaction time at room temperature were observed by using a different transfer ligand.

2P45: Particle size measurement for radioactive aerosols released from pneumatic tubes in research reactor facilities

Ozaki, A.,[a] Kaneto, T.,[a] Oki, Y.,[b] Takamiya, K.,[b] Shibata, S.,[b] Yamasaki, K.[b] ([a]Grad. School of Eng., Kyoto Univ., [b]Res. Reactor Inst., Kyoto Univ.)

Radioactive aerosols are released from the exits of pneumatic tubes into hot cells when irradiation capsules are transported from the reactor core to the hot cells by the pneumatic transport system in research reactors. In this study, we measured size distributions of radioactive aerosols released from the pneumatic tube (Pn-2) in the Kyoto University Reactor using a low-pressure impactor system. Compressed CO_2 gas is used for transport of irradiation capsules in the pneumatic system. Both of FP nuclides and non-FP nuclides (^{24}Na *etc.*) were collected as aerosols from the exit of the tube. The tube at the reactor core is made of aluminum. FP nuclides were considered to be emitted into the compressed gas by recoil of FPs from uranium contained as impurity in the aluminum tube, while non-FP nuclides are formed by activation of airborne dusts in the tube. Particle diameter for FP nuclide aerosols was found to be 0.3–0.5 μm, which was always smaller than that for non-FP nuclides (*ca.* 1μm).

2P46: Spectra of high energy neutrons at the high energy neutron irradiation facility of KENS

Matsumura, H.[a], Masumoto, K.[a], Nakao, N.[a], Wang, Q.[a], Toyoda, A.[a], Kawai, M.[a], Aze, T.[b], Fujimura, M.[b] ([a]KEK, [b]Nihon Univ.)

In high-energy proton-accelerator facilities, generated secondary neutrons should be considered in the viewpoint of radiation safety. For the design of shielding for the neutrons, we have to know the spectra and intensity of the neutrons passing through the shield concretes. However, there are only a few experiments that investigated about them. In this study we investigated the spectra of high-energy-neutrons of 0 degree direction generated with the primary 500MeV protons at the high-energy-neutron irradiation facility of KENS by the means of the radiochemical technique. Some kinds of metallic foils were used as target. The targets were put on 0, 40, 80, and 130cm of depth from the surface of the shield concrete. After irradiation, the yields of spallation products on the targets were determined by gamma-ray spectrometry. The results indicate that the high-energy part of the obtained neutron spectrum is much lower than that estimated by MARS14 code which is the Monte Carlo simulation code.

2P47: Radiation streaming experiment through a labyrinth of the high energy accelerator facility

Nakamura, H.,[a] Miura, T.,[a] Numajiri, M.,[a] Nakao, N.,[a] Nakashima, H.,[b] Matsuda, N.,[b] Watanabe, S.[c] ([a]KEK, [b]JAERI, [c]Tokyo Nuclear Service)

It is important to evaluate radiation streaming through a labyrinth of accelerators for radiation shield design. There are several calculations for evaluation. However these methods have not been tested sufficiently at high energy accelerators. In this experiment we used a cupper target inserted in the neutrino beam line of the 12GeV proton accelerator facility as a radiation source, measured radiation dose along the labyrinth of the facility by using radiation dosimeters and activation detectors, compared the measurement with Monte Carlo methods. This time we report about the measurement by TLDs only. We used TLDs (thermo luminescence dosimeter, Panasonic UD813PQ4) and the moderated cases (Panasonic UDS893P-1) which moderates fast neutron to thermal neutron. The luminescent materials of the TLD are two kinds of lithium borates ($^6Li_2{}^{10}B_4O_7$(Cu) and $^7Li_2{}^{11}B_4O_7$(Cu)) which respond to thermal neutron differently. The difference of the respondency with the moderated case shows neutron dose. The measurement data were compared with one calculated by Monte Carlo code; MCNPX. At the labyrinth far from the beam line measured ones were smaller than calculated ones. In contrast, at the labyrinth near from the beam line measured ones were larger than calculated ones.

2P48: γ-ray spectrometry by well-type Ge-detector using ^{176}Lu as internal standard

Takamiya, K.,[1] Sasaki. T.,[2] Goto, S.[3] ([1]Res. Reactor Inst., Kyoto Univ., [2]Faculty of Engineering, Kyoto Univ., [3]Faculty of Science, Niigata Univ.)

Well-type Ge-detectors are usually used for measurement of radioactivities of environmental materials and other samples that has weak radioactivities because of their high detecting efficiencies. But it is difficult to determine the radioactivities of the samples correctly. The method of determining radioactivities by well-type Ge-detector using ^{176}Lu as internal standard has been developed. ^{176}Lu decays with emission of gamma rays at low energy region (88, 202, and 307 keV), and detection efficiency of the detector could be calculated by photo peaks of these gamma rays. In order to examine the coincidence summing effect and detection limit of this method, measurements for Lu_2O_3 of 1 and 10 mg were carried out. Photo peaks of gamma rays of ^{176}Lu and their sum peaks were observed definitely. The analysis method taking account of coincidence summing effect using these photo peaks will be developed and applied for environmental standard samples.

2P49: Determination of ^{240}Pu/^{239}Pu ratios in soil related to its chemical species

Muramatsu, Y.[a], Yoshida, S.[a], Yamazaki, S.[b] ([a]National Institute of Radiological Sciences, [b]Tokyo Nuclear Service)

We have determined Pu concentrations and the ^{240}Pu/^{239}Pu ratios in soil samples collected from different places in Japan by HR-ICP-MS. The ratios obtained for about 20 Japanese soils were in the range 0.155 – 0.194 (average: 0.180 +/- 0.011). In order to obtain information on the chemical species of Pu in soil, we have extracted this element by sequential extraction method. For this we

used different concentrations of nitric acid to extract Pu from soil. Then the residue was decomposed by fluoric acid, followed by alkaline fusion. It was found that the $^{240}Pu/^{239}Pu$ ratio in the first fraction extracted by diluted nitric acid (e.g. 0.3N) was markedly higher than that in the concentrated one. The $^{240}Pu/^{239}Pu$ ratios in the residues, which were decomposed by fluoric acid and also by alkaline fusion, were significantly lower (down to about 0.05) than the bulk value (0.17). These indicate that there are different Pu species (or fractions) with different $^{240}Pu/^{239}Pu$ ratios in soil.