English Abstracts

1A01 Studies on hot atom chemical behavior of energetic ions in solids1 (XIX) -Dependence of implantation ion fluence on chemical behavior of energetic hydrogen isotopes implanted into carbon-contained boron films-

OSUO, J.[1], SUZUKI, S.[1], KOBAYASHI, M., KURATA, R.[1], WANG, W.[1], ASHIKAWA, N.[2], SAGARA, A.[2], OYA, Y.[1], OKUNO, K.[1]
([1]Radiosci. Res. Lab. Fac. of Sci., Shizuoka Univ., [2]NIFS)

For the first wall of D-T fusion reactor, boronization has been thought to be applied for the reduction of impurities such as oxygen and carbon. In the fusion environment, energetic particles including tritium would be implanted into the boron film and trapped with various chemical states. For the estimation of tritium retention, it is important to understand the interaction of tritium chemical behaviors not only with boron but also with impurities in boron film. In this study, our attention was paid to the role of carbon on tritium trapping processes in boron films.

A carbon–contained boron film with the carbon concentration of 40% was prepared by the plasma CVD apparatus, and deuterium ions (D_2^+) were implanted into the film with various ion fluences. The chemical states of carbon and boron were evaluated by XPS and the deuterium desorption behavior was revealed by TDS.

D_2 TDS spectra showed that the desorption behavior for samples implanted at ion fluences (0.5 -2.0) $\times 10^{22}$ D^+ m^{-2}, consisted of three D_2 desorption stages, namely, the desorption of deuterium bound to boron and carbon with B-D-B, B-D and B-C-D bonds However, for the sample implanted at ion fluences of 0.1 $\times$ 10^{22} D^+ m^{-2}l, only B-C-D bond was formed. These results indicated that the deuterium would be preferentially trapped by carbon compared to boron.

1A02 Studies on hot atom chemical behavior of energetic ions in solids (XX) - Dependence of ion fluence on deuterium retention behavior in simultaneous deuterium and carbon ions implanted tungsten

MATSUOKA, K.[1], SUZUKI, S.[1], KOBAYASHI, M.[1], KURATA, R.[1], WANG, W.[1], ASHIKAWA, N.[2], SAGARA, A.[2], YOSHIDA, N.[3], OYA, Y.[1], OKUNO. K.[1]
([1]Radiosci. Res. Lab. Fac. of Sci., Shizuoka Univ., [2]NIFS, [3]Res. Inst. for App. Mech., Kyushu Univ.)

In D-T fusion reactors, the combination usage of tungsten (W) and carbon (C) has been considered as the divertor materials to endure the high heat flux from the plasma. Under this condition, some of C will be ionized and implanted into W and the W-C mixed layer will be formed. In addition, hydrogen isotopes including tritium will be also implanted into the mixed layer, simultaneously. Therefore, the dynamics of hydrogen isotopes during the formation of the W-C mixed layer should be elucidated for the estimation of tritium inventory. In this study, the dependence of ion fluence on deuterium retention in the simultaneous C^+ and D_2^+ implanted W were investigated by means of TDS and XPS. The D_2 TDS spectra showed that it consited of three D_2 desorption stages The peak around 950 K corresponded to the desorption of deuterium trapped as C-D bond. This peak increased in the lower fluence, whereas that in the higher fluence, it decreased, indicating that the chemical sputtering of deuterium by C would govern the deuterium retention compared to the trapping of deuterium by the C in the higher fluence.

1A03 Studies on hot atom chemical behavior of energetic ions in solids (XXI) -Elucidation of oxidation temperature effect on deuterium retention in the oxide layer of stainless steel-

SUZUKI, M.[1], SUZUKI, S.[1], KOBAYASHI, M.[1], KURATA, R.[1], WANG, W.[1], HAYASHI, T.[2], YAMANISHI, T.[2], OYA, Y.[1], OKUNO, K.[1]
(Radiosci. Res. Lab., Fac. of Sci., Shizuoka Univ.[1], JAEA[2])

In fusion device, it is considered that type 316 stainless steel (SS316) will be widely used as the vacuum chamber, cooling pipes and so on. It is known that the oxide layer formed on the surface will affect the retention behavior of hydrogen isotopes in SS316. In addition, the chemical states of the oxide layer depend on the oxidation temperatures of SS316. In this study, the effect of oxidation temperature on the retention behavior of hydrogen isotopes was studied.

After preheating for the removal of residual oxide layer, the samples were exposed to air for 1 hour at R.T., 473 K and 673 K to form the oxide layers. The D_2^+ was implanted into the sample at R.T. Thereafter, the deuterium desorption and the chemical states of oxide layer were elucidated by TDS and XPS, respectively.

The TDS results showed that the deuterium desorption peak around 600-800 K increased as the oxidation temperature increased. According to the XPS measurements, the major chemical state of the oxide layers was the iron oxide, and the oxygen concentration and the chemical state of iron in the layer increased with increasing the oxidation temperature. These results concluded that the chemical states of iron oxide would affect the deuterium retentions in the oxide layer.

1A04 Studies on hot atom chemical behavior of energetic ions in solids (XXII) – Annihilation behavior of irradiation defects generated in 14 MeV neutron-irradiated lithium aluminate -

HAMADA, A[1]., SUZUKI, S[1]., KOBAYASHI, M[1]., KURATA, R[1]., WANG, W[1]., OCHIAI, K[2]., KONNO, C[2]., OYA, Y[1]., OKUNO, K[1]
(Radiosci. Res. Lab., Fac. of Sci., Shizuoka Univ.[1], JAEA[2])

In blanket systems for the D-T fusion reactor, it is considered that lithium ceramics are introduced to breed tritium by the reactions of ^{6}Li (n, α) T and ^{7}Li (n,n' α) T. Lithium aluminate ($LiAlO_2$) is one of the candidates for tritium breeding materials due to its chemical stability. In the fusion reactors, 14 MeV neutrons were generated by D-T fusion reaction and would impinge on tritium breeding materials. It is expected that tritium generated in the breeding materials would be trapped by irradiation defects induced by 14 MeV neutron-irradiation. Therefore, it is important to understand the correlation between the annihilation behavior of irradiation defects and the tritium release behavior. In this study, the annihilation kinetics of irradiation defects in 14 MeV neutron-irradiated $LiAlO_2$ was elucidated by Electron Spin Resonance (ESR).

From the ESR spectra, F^+-center, O^--center and O_2^--center were formed as irradiation defects. According to the isochronal annealing experiments, it was indicated that F^+-center and O^--center were disappeared by the recombination and F-center aggregation (FA) was formed at the temperature range of 300 - 500 K. Above 500 K, thereafter, FA and O_2^--center began to disappear by the decomposition.

1A05 A proof-of-principle experiment of EIT with gamma radiation in FePSe$_3$ single crystal

NAKAJO, S[1]., SUZUKI, H[2]., NAGATA, K[1].,NAKAGAMI, K[1].,MURAMATSU, H[2].
([1]Grad. School of Edu., Shinshu Univ., [2]Fac of Edu., Shinshu Univ.)

We have already reported a proof-of-principle experiment, demonstrating electromagnetically induced transparency (EIT) with gamma radiation for the Mössbauer effect using the level-crossing technique. In this report, we will discuss a possibility of EIT on FePSe$_3$ single crystal. We synthesized a single crystal of FePSe$_3$ by solid phase reaction in the vacuum-shielded tube (Vycol glass tube) including the mixture of stoichiometric amounts of Fe, P, Se at 840℃ for about 1 month. The absorber FePSe$_3$ is antiferromagnetic below the Néel temperature of 107K, which means that the hyperfine field can be varied by changing the temperature. A strong temperature-dependent internal magnetic field parallel to the EFG-axis in the single crystal allows for crossings in the nuclear level structure of ^{57}Fe. From the analyses of a series of Mössbauer spectra taken at various temperatures, we determine that at 27 K the states $|I=3/2, m=-3/2\rangle$ and $|I=3/2, m=1/2\rangle$ cross and are mixed. Two of the absorption lines ($m=-1/2 \leftrightarrow m=-3/2$) and ($m=-1/2 \leftrightarrow m=1/2$) merge into one single line. So, we can see some deficit of absorption in this sample.

1A06 U-238 Mössbauer spectroscopic study of uranium oxysulfide

MASAKI, N.[1], NAKADA, M.[1], AKABORI, M.[1], ARAI, Y.[1], NAKAMURA, A.[1], SATO, N.[2]
([1]Japan Atomic Energy Agency, [2]Inst. of Multidisciplinary Research for Advanced Materials, Tohoku Univ.)

The U-238 Mössbauer effect of uranium oxysulfide, UOS, has been investigated at various temperatures from 19 K to 70 K. The γ-ray source used for U-238 Mössbauer spectroscopy was Pu-242 dioxide with high isotope purity. By using a pure Ge detector, the 44.9 keV U-238 Mössbauer γ-ray is easily distinguished from the 59.5 keV γ-ray of the Am-241 isotope, which is the daughter nucleus of the isotopic impurity Pu-241. Quadrupole splitting of the U-238 nucleus in UOS is approximately 40 mm/s in the temperature range without account of magnetic splitting. According to neutron diffraction and magnetic susceptibility measurements, UOS is antiferromagnetic at low temperatures with a Neel temperature of T_N = 55 K. However, Mössbauer spectra below 55 K could be analyzed by using only quadrupole splitting without magnetic splitting. This result showed that the magnitude of quadrupole splitting was much larger than that of magnetic splitting of U nucleus in UOS.

1A07 Crystal structures, and ^{151}Eu and ^{155}Gd Mössbauer spectra of lanthanide complexes with phosphine oxides

TAKAHASHI, M., HIRAI, Y.
(Department of Chemistry, Toho Univ.)

The lanthanide complexes having phosphine oxides such as triphenylphosphine oxide (TPPO) and triethylphosphine oxide (TEPO) are investigated. The TPPO complexes Ln (tppo)$_4$ (ClO$_4$)$_3$ · MeOH (A; Ln = Nd, Eu, Gd, Dy, Yb) are isostructural and contain dodecahedral [Ln (tppo)$_4$ (ClO$_4$)$_2$]$^+$ ion having two chelating ClO$_4^-$ ions. The coordination of ClO$_4^-$ in a methanol solution is also suggested from ESI-MS spectrum. The same structures are also found in [Ln (tepo)$_4$ (ClO$_4$)$_2$] ClO$_4$. (B) The nitrato complexes contains nine-coordinate [Ln (tppo)$_2$ (NO$_3$)$_3$ (EtOH)] (C; Ln = Eu, Gd). The europium (III) complexes of A–C show strong emission due to $^7F_n \leftarrow {}^5D_0$ transitions. A detail analysis of the emission spectra suggests that the local symmetry of Eu–C is higher than those of Eu-A and Eu-B. The same is also suggested from the ^{151}Eu and ^{155}Gd Mössbauer spectra; The electric field gradient V_{zz} values, calculated from the observed quadrupole coupling constants, of Eu-A and Gd-A (3.37×10^{21} and 3.66×10^{21} V m^{-2}) are obviously larger than those of Eu-C and Gd-C (2.32×10^{21} and 1.10×10^{21} V m^{-2}).

1A08 Structural and kinetic investigations for the labeling of biomolecules with the {TcO$_3$}-core

Y. Tooyama, H. Braband and Roger Alberto
(Institute of Inorganic Chemistry, University of Zürich.)

The reactions of the [Tc (VII) O$_3$] complexes with the alkenes give corresponding Tc (V) glycolate complexes. Theses complexes are fully characterized by the ^{1}H-NMR, IR and X-ray analysis. The reaction mechanism was kinetically investigated by following UV-Vis spectrum.

1A09 Accelerator Mass Spectrometry and isotope system in earth environment

MATSUZAKI, H.
(School of Eng., Univ. of Tokyo)

AMS (Accelerator Mass Spectrometry) is an extremely high sensitive mass spectrometry and advantageous especially in detection of long-lived radioisotopes such as ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{129}I. Most important source of these isotopes is the interaction between cosmic ray and the materials on the earth surface. CRNs (Cosmogenic Radio Nuclides) produced in the atmosphere (which are called “meteoric” CRNs) are carried by the material circulation and distributed to every site on the earth surface and deposited. The production rate of the CRNs depends on primarily the cosmic ray intensity which is in turn affected by the geomagnetic field intensity and the solar activity. The solar activity and the geomagnetic field should be strongly related to the global climate and environment. So we can investigate the history of the palaeo environment by analyzing CRNs in the natural archives such as tree rings, corals, speleothems sediments and ice cores. Such palaeo environmental study is one of important application field of AMS as well as radiocarbon dating.

Recently, in situ CRNs (which are produced by cosmic ray directly in the earth surface rocks) are analyzed for the investigation of the earth surface process. The concentrations of in situ CRNs, typically ^{10}Be and ^{26}Al in the quartz, tell us the surface exposure age or the erosion rate of the surface. These data are used not only for the local history of the surface morphology but also for the key information for the regional climate condition.

^{14}C, ^{36}Cl and ^{129}I are also produced anthropogenic processes such as the nuclear weapons testing and the nuclear fuel reprocessing. These nuclides are also potential new tracers for the material dynamics study. Today ^{129}I, for example, concentration level in the earth surface environment has been raised up to several orders higher by the anthropogenic origin. And ^{129}I distribution is fairly different from region to region according to the surface condition and the iodine dynamics in the environment. Elemental iodine is associated with the organic materials. Thus ^{129}I can be not only a good proxy for the elemental iodine dynamics but also the potential clue to reveal a new aspect of the carbon dynamics.

1A10 Accelerator mass spectrometry of ^{36}Cl and its applications

SASA, K.
(Tandem Accel. Complex, Univ. Tsukuba)

Cosmogenic ^{36}Cl is a long-lived radionuclide with a half-life of 301 kyr. It is mainly produced in the stratosphere through spallation of ^{40}Ar by energetic primary and secondary cosmic rays. The global mean production rate of ^{36}Cl has been estimated to be ~20 atoms m^{-2} s^{-1}. Natural ^{36}Cl is transported in about 2 years from the upper atmosphere to the ground and is accumulated in terrestrial deposits. The ^{36}S isobar is the principal challenge confronting AMS measurement of ^{36}Cl. Due to the effective separation between ^{36}Cl and ^{36}S, the higher energies available from the large tandem accelerators are required for ^{36}Cl-AMS. ^{36}Cl is measured routinely several laboratories using the large tandem accelerator. ^{36}Cl is effectively used as a monitoring tool for earth and environmental sciences. Especially, ^{36}Cl is the principal AMS isotope used in hydrology. A large excess of ^{36}Cl in the atmosphere was produced by nuclear weapons testing in the 1950's. This bomb pulse has proved to be a useful tracer for modern water movement. In this presentation, I will introduce the status of ^{36}Cl-AMS, and its applications for earth and environmental sciences.

1A11 Analysis of I-129 and its application to environmental sciences and geochemistry

MURAMATSU, Y.[1], ITO, E., TOYAMA, C.[1], MATSUZAKI, H.[2]
([1]Faculty of Science, Gakushuin Univ.[1], MALT, Univ. of Tokyo[2])

Analytical procedures in the determination of iodine-129 (half-life: 1.6x10^7 y) in environmental materials (e.g. soil, underground brine, rain and seawater) have been studied using accelerator mass spectrometry (AMS). For solid samples, iodine was volatilized by pyrohydrolysis at 1000℃ and collected in an alkaline trap solution. Iodine was purified from the matrix by solvent extraction. For brine samples with high iodine concentrations, samples were directly processed by solvent extraction to purify iodine. Finally, the obtained iodine fractions were precipitated as silver iodide to make targets for AMS at MALT. We have also analyzed stable iodine (^{127}I) concentrations in the samples by ICP-MS. We could determine the ^{129}I/^{127}I ratios down to 1x10-13 in brine samples. The detection limit for soil sample (1 g material) by the present method was about 0.01 mBq/kg or 1x10^{-11} as the ratio of stable iodine (^{129}I/^{127}I ratio), i.e. this value was at least 2 orders of magnitude better than that by neutron activation analysis.

1A12 Archaeology and ^{14}C: present status and issues

IMAMURA, M.[1], SAKAMOTO, M.[1], OZAKI, H.[2], MIYATA, Y.[3]
([1]National Museum of Japanese History, National Institute for Humanities, [2]Paleo Labo Inc., [3]Center for Chronological Research, Nagoya Univ.)

Recent advance in ^{14}C accelerator mass spectrometry has greatly improved its performances, enabling routine ^{14}C measurements of a few thousands samples per year at the precision levels of 0.2~0.4 % using a small accelerator. This, along with the calibration techniques using the universal atmospheric ^{14}C dataset, has made radiocarbon dating much more powerful tool in archaeological applications than we saw before 2000. In this paper authors review the present status in methods of precision dating and the outcome of some ^{14}C applications mostly based on our research practices performed in the last several years. The related issues are also discussed on problems such as sample selection and calibration, in prospect of future improvements. Particularly, the ^{14}C regional effects with orders of 0.5% or more found by our group in Japanese wood poses the necessity of detailed calibration data for the region of interest. The phenomena are seemingly correlated with the climatic change, and may provide an interesting information if compared with other climate proxies.

1A13 Correlation between the concentrations of cosmogenic Be isotopes in atmosphere and solar activities.

YAMAGATA, T.[1], NARAZAKI, Y.[2], IKEDA,Y.[3], H. NAGAI, H.[3], MATSUZAKI, H.[4]
([1]Grad. School of Integ. Basic Sci., Nihon Univ., [2]Fukuoka Inst. of Health and Environ. Sci., [3]College of Human. and Sci., Nihon Univ., Sch. of Engin., Univ. of Tokyo)

The concentrations of 7Be and 10Be were investigated at Dazaifu, Fukuoka (1998-2002), Hachijo-Island (2002-2005) and Tokyo (2002-2008) during 1998 to 2008. The seasonal variations were same each year; high concentrations and high isotopic ratios of 10Be/7Be that was caused by strong stratosphere-troposphere exchange (STE) were appeared in February to June, and various concentrations but constant 10Be/7Be by vertical convection in troposphere were appeared in July to December. The concentrations were reconstructed by the box model formula. The parameters of the mean residence time and STE intensities, and period, were constant. The amplitudes of production rate were higher than the amplitude of cosmic ray neutron flux observed at Moscow near earth's surface by a factor of 4. Since the neutron flux amplitude at polar region that was little influenced by geomagnetic field was only 10% higher than Moscow, the high amplitudes of production rate were assumed that caused by changing of energy spectrum of galactic cosmic ray.

1A14 Isotope ratios of ^{36}Cl/Cl in soils by AMS

TAMARI, M.[1], SUEKI, K.[1], AMANO, T.[1], SASA, K.[1], TAKAHASHI, T.[2], MATSUSHI, Y.[3], TOSAKI, Y.[2], OKI, T.[2], NAGASHIMA, Y.[2], KINOSHITA, T.[2], MATSUMURA, H.[4], BESSHO, K.[4]
([1]Grad. School of Pure and Applied Sci., Univ. of Tsukuba, [2]AMS Group, Univ. of Tsukuba, [3]Micro Anal. Labo., Tandem Accel., the Univ. of Tokyo, [4]High Ener. Accel. Res. Organ.)

We measured ^{36}Cl/Cl ratios of soil samples collected at the equal-latitude cross-sectional areas (37° 20' N - 37° 30' N) in Japan. ^{36}Cl/Cl depth profiles were also obtained for three sites. We developed an improved leaching process that uses diluted HNO_3 as an extractant, activated carbon to remove organic matters without decomposition, and H_2O_2 to remove residual organic matters. This sequential treatment allows us to measure selectively ^{36}Cl/Cl ratios of inorganic chlorine in soil. The isotope ratios ^{36}Cl/Cl of soil samples were determined by accelerator mass spectrometry (AMS) at the Tandem accelerator, University of Tsukuba. We have collected 27 surface soil samples from 6 sites at the equal-latitude cross-sectional areas. The measured ^{36}Cl/Cl ratios of these samples were between 0.1 x 10^{-13} and

3.7 x 10^{-13}. Surface soil $^{36}Cl/Cl$ depth profiles measured at KK-12 and AB-1 are shown in Figure. The depth profiles showed that the $^{36}Cl/Cl$ ratio value had higher peak at a shallow point (between 0 - 15cm). The $^{36}Cl/Cl$ ratio of AB-1 (peak value was 3.7 x 10^{-13}) was two times of KK-12 (peak value was 2 x 10^{-13}). In the KK-12 from 15 to 100 cm depth, the $^{36}Cl/Cl$ ratios drop to become observed $^{36}Cl/Cl$ ratio in snow at KK site and predicted $^{36}Cl/Cl$ ratio. But, in the AB-1 below 30 cm depth, the $^{36}Cl/Cl$ ratios were flat at 1 x 10^{-13} and were three - four times of predicted value.

1A15 The distribution of ^{129}I in the Japan Basin and Yamato Basin

SUZUKI, T.[1], MINAKAWA, M.[2], TOGAWA, O.[1]
([1]Research Group for Environmental Science, JAEA, [2]Radioecology Section, NRIFS)

Because of the results form the decrease of dissolved oxygen and increase of seawater temperature, the Japan Sea is known as a sensitive ocean for global warming. To investigate water circulation in the Japan Sea bottom water (JSBW), ^{129}I and ^{14}C in seawater samples were measured by accelerator mass spectrometry at the Aomori Research and Development Center, Japan Atomic Energy Agency. The $^{129}I/^{127}I$ ratios at the JSBW in the Japan Basin and Yamato Basin were $(6.7 \pm 0.1) \times 10^{-12}$ and $(9.2 \pm 0.1) \times 10^{-12}$, respectively. The difference of the ratio between two basins would result from the concentration of ^{129}I in surface seawater when the JSBW was formed by winter convection. Therefore, we concluded that ^{129}I would be one of the most useful tracers to investigate the oceanic circulation in the Japan Sea.

1A16 Production rate of ^{36}Cl in chondritic meteorites (II)

HAMANAKA, Y.[1], OURA, Y.[1], EBIHARA, M.[1], SASA, K.[2], NAGASHIMA, Y.[2], TAKAHASHI, T.[2], TOSAKI, Y.[2], MATSUSHI, Y.[3], TAMARI, M.[2], AMANO,T.[2], SUEKI,K.[2], BESSHO, K.[4], KINOSHITA, N.[2]
([1]Grad. School of Sci. Eng., Tokyo Metropolitan Univ., [2]AMS Group, Univ. of Tsukuba, [3]School Eng. Univ. of Tokyo, [4]KEK)

^{36}Cl ($T_{1/2}$ = 3.01 × 10^5 y) is a one of the cosmogenic nuclides, and produced by cosmic ray induced reaction such as spallation reaction and neutron capture reaction of ^{35}Cl. Excitation functions of ^{36}Cl production by proton-induced spallation reaction are characteristic for target elements. Thus, elemental production rates of ^{36}Cl should provide additional cosmochemical information on radiation history. We tried to estimate experimentally an elemental production rate of ^{36}Cl by spallation reactions as well as neutron capture reaction in two chondritic meteorites. Elemental concentrations and ^{36}Cl concentration in a bulk sample and 5 separated phases from two fragments of Gold Basin meteorite and a fragment of Gao meteorite were determined by 3 kinds of activation analysis and accelerated mass spectrometry, respectively. For all fragments, silicate phases dissolved in HNO_3 had highest both ^{36}Cl concentration and Cl elemental concentration among 5 phases. It suggests that the contribution of Cl to ^{36}Cl is large. The production rates of ^{36}Cl for main target element groups, Cl, (K + Ca) and (Fe + Ni), were calculated from ^{36}Cl and target elemental concentrations for each fragment. Obtained production rates in this study were nearly consistent with those from model calculation by Welten et al. Our way for estimation of production rates is proved to be reasonable.

1A17 AMS measurement of p-nuclei Sm-146

KINOSHITA, N[1]., PAUL, M[2]., FERINBERG, G[2]., NASSAR, H[2]., COLLON, P[3]., KASHIV, Y[3]., ROBERTSON, D[3]., SCHMITT, C[3]., TANG, X.D[3]., DEIBEL, C[4]., DIGIOVINE, B[4]., GREENE, J.P[4]., HENDERSON, D.J[4]., JIANG, C.L[4]., KAY, B[4]., LEE, H.Y[4]., MARLEY, S.T[4]., PARDO, R.C[4]., PATEL, N[4]., REHM, K.E[4]., SCOTT, R[4]., VONDRASEK, R[4]., NOTANI, M[4]., FIGUEIRA, J.M[5]., GORIELY, S[6]., JISONNA, L[7]., AMAKAWA, H[8]., TAKAHASHI, N[9]., MITSUGASHIRA, T[10]., OHTSUKI, T[11]., HIROSE, K[11]., NAKANISHI, T[12]., YOKOYAMA, A[12].
([1]Accelerator Complex, Univ. of Tsukuba, [2]Hebrew Univ., [3]Univ. of Notre Dame, [4]ANL, [5]Tandar Lab., [6]Univ. Libre de Bruxelles, [7]Northwestern Univ., [8]Ocean Res. Inst., Univ. of Tokyo, [9]Grad. School of Sci., Osaka Univ., [10]Inst. of Multidiciplinary Res. for Advanced Materials, Tohoku Univ., [11]Lab of Nucl. Sci., Tohoku Univ., [12]Inst. of Sci. and Eng., Kanazawa Univ.)

The ^{146}Sm nuclide belongs to the small family of nuclides produced by *p*-process of nucleosynthesis. Its presence in the early Solar System was established by the study of isotopic anomalies of its α-daughter, ^{142}Nd, in meteorites. What is very interesting is a difference in the ^{142}Nd isotopic signatures between terrestrial and chondritic materials, leading to a new insight in the chronology of planetary formation. We have developed a method for the ^{146}Sm detection by positive-ion high-energy accelerator mass spectrometry (AMS) with a gas-filled magnet for the isobaric ^{146}Sm-^{146}Nd separation at the Argonne ATLAS facility. The ^{146}Sm samples were produced in (γ, n), (n, 2n) and (p, 2nε) activations of enriched ^{147}Sm targets. A new determination of the ^{146}Sm half-life in progress is based on both measurements of the α activity ratios of $^{146}Sm/^{147}Sm$ in the ^{146}Sm samples and the $^{146}Sm/^{147}Sm$ atomic ratios of the same samples by AMS. Special attention is given to determination of the latter ratios in the present study.

1A18 On Possibility of Standardization of Analysis Method of "Bio-Degree" of the Biomass-based Materials

K. OHSHIMA[1]., T. KIMURA[2], K. KOSHIISHI[1]
([1]Bioindustry Development, Japan Bioindustry Association, [2]Division of Bioresources and Product Science, Grad. School of Agriculture Research, Faculty of Agriculture, Hokkaido Univ.)

Possibility of the standardization on analysis method of "Bio-Degree", content of the component derived from the biomass in materials, based on the measurement of content of ^{14}C by AMS[a)] and LSC[b)], was experimentally investigated.

The "Bio-Degree" of about 30 materials derived from various type of the biomass was measured and compared with the theoretical content. For the biofuels, that are bio-ethanol or bio-ETBE[c)] containing gasoline or BDF[d)], direct LSC method, counting directly the ^{14}C scintillation from the specimen without the material conversion, different from Method A[e)] or C[f)] of ASTM D6866, was seemed to be available for the standardization, whereas for solid materials like as bio-based polymers, Method B[e)] of ASTM D6866 or its advanced preparation method, where graphite is formed as specimen for AMS, was most reasonable for the standardization. Based on these positive results, most basic standardization conditions for JIS/ISO registration on the direct method of LSC for the biofuels are now investigated in Japan Standard Association's programs.

(*a) AMS:Accelerated Mass Spectroscopy
(*b) LSC:Liquid Scintillation Counting
(*c) ETBE: Ethyl Tertiary-Butyl Ether
(*d) BDF:Bio-Diesel Fuels
(*e) Method A/B of ASTM D6866:Sample is converted to CO_2 for LSC/AMS measurement
(*f) Method C of ASTM D6866: Sample is converted to C_6H_6 for LSC measurement

1B01 Application of the rapid α source preparation method, samarium hydroxide coprecipitation method, to superheavy element chemistry

KASAMATSU, Y., HABA, H., EZAKI, Y.
(Nishina Center, RIKEN)

We established a rapid preparation method of samarium hydroxide coprecipitation samples using multitracer. The multitracer was produced by the nuclear spallation reaction of Ta and transported from the irradiation chamber to the chemistry laboratory by a He/KCl gas-jet system. By the present method, we were able to rapidly prepare the precipitate samples with high energy resolution in the α spectrometry. Furthermore, different chemical yields of many elements depending on their own chemical properties were also obtained. These results indicate that unknown precipitation properties of short-lived superheavy elements would be investigated by the present method. For the investigation of the hydroxide precipitation of element-104 Rf (half-life: 68 s), we also studied hydroxide coprecipitation behavior of Hf and Zr which are the homologues of Rf with radiotracers of ^{88}Zr and ^{175}Hf.

1B02 On-line isothermal gas chromatographic behavior of group 5 elements Nb and Ta as homologues of Db

SATO, T. K.[1], TSUKADA, K.[1] , ASAI, M.1 , TOYOSHIMA, A.[1] , KASAMATSU, Y.[2] , LI, Z.[1] , SATO, N.[1] , KIKUCHI, T.[1] , NAGAME, Y.[1]
([1]Japan Atomic Energy Agency, [2]Nishina Center for Accelerator Based Science, RIKEN)

To investigate chemical properties of the transactinide element dubnium (Db, Z=105), we have developed an on-line isothermal gas chromatographic apparatus. The on-line experiments with the group-5 elements using the short lived ^{88}Nb and ^{170}Ta as the homologues of Db were conducted. We measured the relative chemical yields of volatile compounds of ^{88}Nb and ^{170}Ta as a function of the isothermal temperature in chlorinating condition with mixture of air and the $SOCl_2$ vapor. It was found that the volatility of compounds of Ta is lower than that of Nb.

1B03 Research on transportation of chemical compounds after gas phase chemistry

MURAYAMA, H.[1],GOTO, S.[1], KUDO, H.1,TUKADA, K.[2], ASAI, M.[2],TOYOSIMA, A.[2], SATO, T.[2],NAGAME, Y.[2]
([1]Fac.of Sci.,Niigata Univ., [2]JAEA.)

A gas-phase chemical separation method is one of the most useful techniques to investigate chemical properties of super heavy elements, which examines behavior of adsorption and desorption of volatile compounds on a column surface. Our group has performed the gas-phase experiments for chlorides of group-4 elements (Zr,Hf,and Rf). Because the super heavy elements such as rutherfordium are mostly short-lived nuclides and alpha emitters, they have to be re-transported quickly and efficiently to an alpha detection site after passing through the column. However, there are few reports about the conditions of re-transportation for the volatile compounds of super heavy elements. In this study, we constructed an apparatus for re-transportation of volatile compounds and searched optimum the re-transportation conditions.

The experiments were performed using fission products from ^{252}Cf. The re-transportation efficiency for the volatile compounds of ^{101}Tc,^{104}Tc, and ^{107}Rh which were observed clearly at gamma spectrum reached about 40%, when KCl aerosol and N_2 carrier gas were used. The detailed results, for example dependences on kinds of carrier gas, an aerosol, and so on, will be reported.

1B04 Application of laser ionization mass spectroscopy for gas phase chemistry

SHIGENO, Y. [1] GOTO, S.[2] KUDO, H,[3]
([1]Graduate School of Science and Technology, Niigata Univ., [2]Center for Instrumental Analysis Niigata Univ., [3]Department of Chemistry, Faculty of Science, Niigata Univ.)

In order to study chemical properties of transactinoid elements it is necessary to apply a rapid chemistry such as a gas phase chemistry. The gas phase chemistry is based on volatility of complexes of the objective elements. At a preliminary experiment, it was found that several elements showed a volatility as dipyvaloylmethane (DPM) complexes. However, their chemical forms have not been made clear yet. Therefore, the mass spectrometer has been used for elucidation of their chemical forms. There are two requirements for an ionization of mass spectroscopy. First requirement is that nondestructive molecular ion peak showed be observed and the second is high-selectivity. One of he ionization method to satisfy these requirements is Resonance Enhanced Multi-Photon Ionization (REMPI) method.

In order to clarify species of ions generated with REMPI the ion source of commercial mass spectrometer was modified. In the mass spectrometry of DPM with REMPI, intense m/z= 57 and 127 (these are the fragment ions of DPM) were observed. The dependence of the intensity of various ion on laser wavelength and power will be reported.

1B05 Anion-exchange behavior of element-105 Db in HF/HNO_3 mixed solution

KASAMATSU, Y.[1,2], TOYOSHIMA, A.[1], ASAI, M.[1], TSUKADA, K.[1], LI, Z.[1], ISHII, Y.[1], SATO, T. K.[1], NISHINAKA, I.[1], KIKUCHI, T.[1], HABA, H.[2], KUDOU, Y.[2], SATO, N.[1,2], OURA, Y.[3], AKIYAMA, K.[3], OOE, K.[4], FUJISAWA, H.[4], SHINOHARA, A.[4], GOTO, S.[5], KUDO, H.[5], ARAKI, M.[6], NISHIKAWA, M.[6], YOKOYAMA, A.[7], NAGAME, Y.[1]
([1]Advanced Sci. Res. Center, JAEA, [2]Nishina Center, RIKEN, [3]Grad. School of Sci. and Eng., Tokyo Metropolitan Univ., [4]Grad. School of Sci., Osaka Univ., [5]Fac. Sci., Niigata Univ., [6]Grad. School of Nat. Sci. and Tech., Kanazawa Univ., [7]Inst. Sci. and Eng., Kanazawa Univ.)

Dubnium-262 was produced in the ^{248}Cm (^{19}F, 5n) ^{262}Db reaction at the JAEA tandem accelerator. The reaction products were rapidly transported by a He/KF gas-jet system to the chemistry laboratory and the anion-exchange behavior of Db in HF/HNO_3 solution was investigated with a newly developed on-line ion-exchange and α-particle detection apparatus. It was found that distribution coefficients of Db were clearly smaller than those of Ta, the closest homologue in the periodic table, and were close to those of the lighter homologue Nb and also the pseudo homologue Pa. From the present results, the fluoro complex formation of Db will be discussed in the conference.

1B06 Study on the reversed phase chromatography with TTA or TIOA extractant for chemistry of Element 104, rutherfordium (Rf)

ARAKI, M.,[1] TAKEDA, Y.,[2] NANRI, T.,[1] YOKOYAMA, A.[3]
(Grad. School Nat. Sci. Tech., Kanazawa Univ.,[1] Fac. Sci. Kanazawa Univ.,[2] Inst. Sci. Eng., Kanazawa Univ.[3])

In order to investigate a chemical property of Rf, a transactinide of group 4, we prepared resin containing TIOA extractant or TTA extractant for reversed-phase chromatography, and studied solvent extraction behaviors of its homologues, Zr and Hf with the resins to confirm whether they can be applied for the Rf experiment. The batch extraction experiment was performed to measure the distribution ratios (*D*) by using tracers of ^{88}Zr and ^{175}Hf. The off-line column experiment was performed with a macro-size column, a micro-column, and teflon tube. Measured *D* values with the column method with macro-size columns agree with those by the batch method. In the micro-column experiment, obtained percent adsorptions reflect the *D* values by the batch method, but the *D* values cannot be determined from the experiment due to incomplete equilibration. We need to explore the conditions which increase the rate of reaction and / or theoretical methods leading to determination of *D* values under the conditions.

1B07 Extraction behavior of molybdenum as homologue of element 106, seaborgium

OOE, K.[1], YAHAGI, W.[1], KOMORI, Y.[1], FUJISAWA, H.[1], TAKAYAMA, R.[1], KIKUNAGA, H.[1], YOSHIMURA, T.[1], TAKAHASHI, N.[1], HABA, H.[2], KUDOU, Y.[2], EZAKI, Y.[2], TAKAHISA, K.[3], SHINOHARA, A.[1]
([1]Graduate School of Science, Osaka University, [2]Nishina Center for Accelerator-Based Science, RIKEN, [3]Research Center for Nuclear Physics, Osaka University)

Last year, we reported the extraction behavior of tungsten as a homologue of element 106, seaborgium (Sg). In this work, the extraction behavior of molybdenum (Mo), which is also the lighter homologue of Sg, was investigated with the same extraction system. A carrier-free isotope, ^{90}Mo ($T_{1/2} = 5.7$ h), was produced in the ^{nat}Ge (^{22}Ne, xn)^{90}Mo reactions using the RIKEN K70 AVF cyclotron. Reaction products recoiling out of the target were transported by a He/KCl gas-jet method to a chemistry laboratory. The transported products were dissolved in 0.1–11 M hydrochloric acid. Equal volume of 0.05 M tetraphenylarsonium chloride (TPAC)-chloroform or 0.05 M Aliquat 336-chloroform solution was mixed with the aqueous phase. The mixture was shaken for 3–15 min. The two phases were taken into separate polypropylene tubes, and were subjected to γ-ray spectrometry using Ge detectors. The distribution ratio (*D*) of Mo increases with an increase of HCl concentration, indicating that the anionic chloride complex of Mo is formed. The dependence of the *D* values of Mo and W on the concentration of extractant (Aliquat 336) was also investigated to examine the extracted species. The results indicate that the net charge of the Mo and W chloride complexes are -1.

1B08 Reduction of element 101, mendelevium, with flow electrolytic column chromatography

TOYOSHIMA, A.[1], TSUKADA, K.[1], ASAI, M.[1], SATO, T. K.[1], LI, Z. J.[1], SATO, N.[1], KIKUCHI, T.[1], KITATSUJI, Y.[1], NAGAME, Y.[1], OOE, K.[2], SHINOHARA, A.[2], KASAMATSU, Y.[2], HABA, H.[2]
([1]Japan Atomic Energy Agency, [2]Grad. School of Sci., Osaka Univ., [3]Nishina Center for Accelerator Based Science, RIKEN)

Reduction of element 101, mendelevium (Md), was studied with flow electrolytic chromatography. Mendelevium-255 with a half-life of 27 min was produced in the ^{248}Cm (^{11}B, 4*n*) reaction at the JAEA tandem accelerator. Reaction products transported by a KCl/He gas-jet method to the chemistry laboratory were collected on a chemistry apparatus. After removing KCl with HDEHP chromatography, elution behavior of Md in 0.1 M HCl was investigated with a flow electrolytic column apparatus at the applied potentials between 0 ~ -0.9 V vs. a Ag/AgCl reference electrode. Elution behavior of Md was the same as that of ^{250}Bk^{3+} at 0 V, showing that the stable Md^{3+} was not reduced to the divalent state. At -0.9 V, elution of Md was quite similar to that of Sr^{2+}, demonstrating that Md^{3+} was successfully reduced to Md^{2+}.

1B09 Actinides extraction and complexation properties of new oxygen-nitrogen hetero donor ligand PTA

KOBAYASHI, T., SUZUKI, S., IKEDA-OHNO, A., SHIWAKU, H., OKAMOTO, Y., AKUTSU, K., YAITA, T.
(Quantum Beam Science Directorate, Japan Atomic Energy Agency)

The development of actinides recognition compound is of increasing importance because it concerns separation techniques in nuclear fuel cycle and/or decontamination of radioactive waste. In particular, the separation of trivalent actinides (An^{3+}) from trivalent lanthanides (Ln^{3+}) is very difficult task due to their chemical similarity. In recent years, the effective An^{3+}/Ln^{3+} separations were observed for some aza-aromatic nitrogen donor ligands. However, most nitrogen ligands are not effective in acidic condition due to protonation of nitrogen atoms. On the other hand, hard oxygen donor ligands, which do not concern in protonation, generally show high extractabilities for both An^{3+} and Ln^{3+} even in acidic condition.

To develop the An^{3+}/Ln^{3+} separation reagent, which allows high extractability in acidic condition, the combination of both nitrogen and oxygen donors has been proposed. Bearing this in mind, we have developed novel oxygen-nitrogen hetero donor ligand, N-alkyl-N-phenyl-1,10-phenanthroline-2-carboxamide (PTA), showing high extractability and selectivity for Am^{3+} over Eu^{3+} even in the acid condition. Thus, in this study, we investigated the complexation equilibria and structural character of the PTA complex by UV/vis spectroscopy, X-ray absorption fine structure, and X-ray crystallography. In this presentation, complexation properties and effect of oxygen donor will be discussed based on equilibrium and structural aspects.

1B10 Selective Adsorption of Trivalent Actinides to Activated Carbon

ARISAKA, M., WATANABE, M., KIMURA, T.
(Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency)

Adsorption behaviors of trivalent actinides (An (III)) and lanthanides (Ln (III)) to activated carbons with and without oxidation by concentrated nitric acid were investigated. Both An (III) and Ln (III) was adsorbed to activated carbons in the range from pH 1 to 4, and the distribution coefficients (K_d) of An (III) and Ln (III) increase with an increase of pH. Activated carbon without oxidation exhibits significant selectivity to An (III) compared with Ln (III). On the other hand, the oxidation to activated carbon deteriorates the selectivity to An (III). The depletion by the oxidation was attributed to the increase of functional groups of oxygen-donor, e.g., carboxyl and/or carbonyl groups because these functional groups are unfavorable for separation between An (III) and Ln (III). The selectivity of activated carbon without oxidation would be attributed to the interaction between π-electrons of aromatic ring and the metal ions.

1B11 Rapid reductive preparation of Np (IV) based on the electrocatalytic reaction

KITATSUJI Y.[1], KIMURA T.[1], KIHARA S.[2]
([1]Japan Atomic Energy Agency, [2]Kyoto Institute for Interesting Chemistry)

The behaviors of reduction of NpO_2^+ to Np^{4+} or Np^{3+} by controlled potential electrolysis were studied, and a unique time course of electrolysis current was observed. It was concluded that NpO_2^+ was reduced by two reduction processes at a platinum electrode: the chemical reaction with Np^{3+} produced from Np^{4+} by electrode reaction and the electrocatalytic reduction by adsorbed hydrogen atom on platinum electrode surface. NpO_2^+ is reduced to Np^{4+} more selectively and with smaller overpotential through electrocatalytic reduction process, though long electrolysis time are required to reduce NpO_2^+ completely. To improve the electrolysis efficiency of the preparation of Np^{4+} from NpO_2^+, the platinized platinum electrode was applied to bulk electrolysis. The rapid preparation of Np^{4+} comparable to that of reversible redox reaction such as NpO_2^{2+}/ NpO_2^+ with small overpotential was achieved successfully.

1B12 Development of Nitrogen Compounds: *N*, *N*-Dialkyl-2-(1*H*-benzimidazol-2-yl)-pyridine-6-carboxamide and Elucidation of Their Complexation Toward f-element Ions.

AKUTSU, K[1, 2]., KOBAYASHI, T[1]., SHIWAKU, H[1]., SUZUKI, S[1]., IKEDA-OHNO, A[1]., OKAMOTO, Y[1]., YAITA, T[1, 2].
([1]Japan Atomic Energy Agency. [2]Graduate School of Science and Engineering, Ibaraki University.)

The separation of trivalent actinides from lanthanides is one of important processes for the treatment of high-level radioactive wastes. Therefore, many nitrogen-donor ligands, which show high separation abilities between trivalent actinides and lanthanides, have been reported so far. On these background, new nitrogen-donor ligands, *N*, *N*-dialkyl-2-(1*H*-benzimidazol-2-yl)-pyridine-6-carboxamide (BIZA) derivatives, having a various length of alkyl groups, were synthesized; their complexation behaviors with trivalent actinides and lanthanides were investigated by spectroscopic titration and XAFS measurements. Both the measurements suggested that all the BIZA derivatives formed Eu (III): BIZA = 1:1 and 1:2 complex with Eu (III) in methanol. Regarding the formation constant between Eu (III) and BIZA, BIZA with octyl group (octyl-BIZA) showed one-order smaller value than that with methyl group (methyl-BIZA). Additionally, the average bond distance of the N, O atoms of octyl-BIZA with Eu (III) was longer than that of methyl-BIZA. These differences might be due to a steric hindrance deriving from bulkiness of their substituent. In this presentation, the extraction results will be also presented.

1B13 Local fields in impurity-doped zinc oxide

SATO, W.1, OHKUBO, Y.[2], YOKOYAMA. A.[1], NAKANISHI, T.[1]
([1]Inst. Sci. Eng., Kanazawa Univ., [2]Res. Reactor Inst., Kyoto Univ.)

Zinc oxide (ZnO) is an intrinsic *n*-type semiconductor, and it is known that its conductivity varies with the presence of dilute impurities. It is hence important to investigate how impurity ions function in ZnO matrix. In our previous work, we applied the time-differential perturbed angular correlation (TDPAC) method with the ^{111}Cd (←^{111}In) probe to a study of local fields in 0.5 at.% Indium-doped ZnO, finding unexpected behavior of conduction electrons at the probe site. However, the specific site occupied by the majority (~ 86%) of the In ions has not been determined yet. In order to obtain information on the site, in this work, we examined impurity-concentration dependence on TDPAC spectra for samples doped with 0.05 – 10 at.% In ions. The electric field gradient (EFG) produced at the probe nucleus has wider distribution with higher concentration of doped In ions, as plausibly expected. In the present paper, we discuss the EFGs at the probe sites and their distribution on the basis of the temperature dependence and impurity-concentration dependence of the TDPAC spectra.

1B14 Magnetism and Mossbauer study of SnO_2 films implanted with ^{57}Fe,

NOMURA K.[1], NEMETH Z.[1], and REUTHER H.[2]
([1]School of Engineering. The Univ. of Tokyo, [2] Forschungszentrum Dresden-Rossendorf e.V.)

SnO_2 films containing 0.1% Sb and 3% Sb, implanted 100keV ^{57}Fe ions with $5x10^{16}$ ions/cm^2 into at the substrate temperature of 500oC in vacuum of $1x10^{-5}$ hPa, were characterized by conversion electron Mossbauer spectrometry (CEMS) and Kerr effect measurements. These films showed ferromagnetism at room temperature. The ^{57}Fe implanted SnO_2 (3% Sb) film showed the smaller Kerr effect than the ^{57}Fe implanted SnO_2 (0.1% Sb) film. Two doublets are assigned to paramagnetic Fe^{3+} and Fe^{2+} species and two sextets are assigned to site A and site B of magnetite for as implanted film. The magnetite was produced much on the top layer, whereas paramagnetic Fe^{2+} species were much included in deep layer. ^{57}Fe implanted SnO_2 (0.1% and 3% Sb) films, post-annealed at 400°C for 6 hours, also showed ferromagnetism at room temperature. It was found from CEMS that ferromagnetic maghemite is produced from magnetite with increase of paramagnetic Fe^{3+} and decrease of Fe^{2+} species.

1B15 Mössbauer spectra of new electrically conductive vanadate glass

NISHIDA, T.,[1,2] YASUMITSU, H[1]., ISODA, S.[2]
([1]Grad. School Advanced Sci. Eng., Kinki Univ. [2]Fac. Humanity-oriented Sci. Eng., Kinki Univ.)

Mössbauer spectra of heat-treated *NTA glass*™ shows a step-by-step decrease in Δ from 0.68 to 0.50 mm s^{-1}, indicating a decrease in the distortion of FeO_4 and VO_4 tetrahedra. Structural relaxation of the glass network results in a decrease in the *Ea* for the electrical conduction from 0.38 to 0.13 eV, reflecting a decrease in the energy gap between the donor level and the conduction band

(CB). An increased number of carriers (electrons/polarons) occupying the CB will be associated with the marked increase in the conductivity from MΩcm to only several Ωcm.

1B16 Mössbauer spectroscopic studies of oxalate and malonate iron (III) complexes

KATADA, M., KIKUCHI, R., MOROZUMI, S., AKIYAMA, K.
(Grad. School of Sci. and Eng., Tokyo Metropolitan Univ.)

The relaxation phenomena for several kinds of oxalate and malonate iron (III) complexes were studied by means of Mössbauer spectroscopy. The Spin-spin interactions of alkylammonium tris (oxalato) ferrate (III) and tris (malonato) ferrate (III) complexes (n=12-18) became small with increase in Fe-Fe distance. However, in the case of n=10 for both complexes the line-width became larger than those of longer n-alkylammonium oxalate and malonate iron complexes. Temperature dependences of line-width on decylammonium oxalate iron (III) complex were observed. It means that the spin-lattice interaction was predominant compared with the spin-spin-interaction.

An asymmetric quadrupole doublets was observed in some oxalate bridged dinuclear iron complexes. The asymmetry arise from the Kramers doublet.

1B17 Improvement of signal-to-noise ratio in ^{57}Mn-in-beam Mössbauer Spectroscopy by utilizing a plastic scintillation counter

NAGATOMO, T.[1], KOBAYASHI, Y.[2], KUBO, M.K.[1], YAMADA, Y.[3], MIHARA, M.[4], SATO, W.[5], MIYAZAKI, J.[6], SATO, S.[7], KITAGAWA, A.[7]
([1]International Christian Univ., [2]RIKEN Nishina Center, [3]Tokyo Univ. of Science, [4]Osaka Univ., [4]Kanazawa Univ., [6]Nihon Univ., [7]National Institute of Radiological Sciences)

We have studied unusual exotic chemical states of ^{57}Fe created through β decays of shortly lived ^{57}Mn nuclei ($T_{1/2}$=1.45 m), which were implanted with high energy into $KMnO_4$, graphite (HOPG), solid oxygen and Si semiconductors, by means of the in-beam emission Mössbauer spectroscopy. To detect the Mössbauer resonant absorptions of 14 keV γ rays emitted form the ^{57}Fe nuclei, we employed a parallel-plate avalanche counter (PPAC), which contained a ^{57}Fe-enriched stainless-steel absorber and was mounted on a Mössbauer transducer. However, there still remains a significant problem that β rays emitted from ^{57}Mn nuclei much degrades a signal-to-noise ratio (S/N) of Mössbauer spectra because the PPAC has poor resolution to discriminate conversion electrons produced through the resonant absorptions from the incoming β rays emitted from ^{57}Mn nuclei. In the present work, the Mössbauer measurement on ^{57}Fe implanted into a nonmagnetic metal Al plate (50 mm square, 1 mm thick) was carried out at HIMAC of NIRS with a detection system modified by employing an anticoincidence method where a plastic scintillation counter (0.5 mm thick) was set between the PPAC and the Al sample in order to reject the β-ray signals. And then, we have succeeded to obtain an emission Mössbauer spectrum of ^{57}Fe in Al at room temperature with very large S/N with the anticoincidence method. We will introduce the experimental procedure and discuss the obtained results in detail.

1B18 In-beam Mössbauer spectroscopy of ^{57}Mn implanted into aluminum oxide

NAGATOMO, T.[1], Y. KOBAYASHI, Y.[2], KUBO, M. K.[1], YAMADA, Y.[3], MIHARA, M.[4], SATO, S.[5], KITAGAWA, A.[5], MIYAZAKI, J.[6], SATO, W.[7]
([1]ICU, [2]RIKEN, [3]Tokyo Univ. Sci., [4]Osaka Univ., [5]NIRS, [6]Nihon Univ., [7]Kanazawa Univ.)

Aluminum oxide (Al_2O_3) has been extensively investigated in materials science since its optical property is controllable by the nature and amount of impurities. In spite of the long history of studies, there still remain unresolved problems of the color changes arising from the presence of impurities. The objective of this work is thus to investigate the interacting nature of Fe ions implanted as impurities with the Al_2O_3 matrix by in-beam Mössbauer spectroscopy with the ^{57}Fe (←^{57}Mn) probe. The experiment was carried out using Heavy Ion Medical Accelerator in Chiba. The ^{57}Mn beam was produced by the nuclear projectile fragmentation reaction of ^{58}Fe (500 MeV/u) with a beryllium target. ^{57}Mn nuclei were implanted into an Al_2O_3 polycrystalline disk (50 mm diameter and 5 mm thickness). The 14.4 keV Mössbauer γ rays emitted from ^{57}Fe (←^{57}Mn) were detected by a parallel-plate avalanche counter combined with a plastic scintillation β-veto counter. The Mössbauer spectrum obtained at room temperature was fitted by assuming two different components consisting of a singlet (I.S. = -0.44 mm/s) and a doublet (I.S. = -0.72 mm/s, Q.S. = 1.9 mm/s). The singlet peak is assigned to high-spin Fe^{3+} at the substitutional Al^{3+} site, and the doublet might be interstitial Fe^{2+}.

1B19 In-beam Mössbauer spectroscopy of ^{57}Mn implanted into magnesium oxide

NAGATOMO, T.[1], KOBAYASHI, Y.[2], KUBO, M. K.[1], YAMADA, Y.[3], MIHARA, M.[4], SATO, W.[5], MIYAZAKI, J.[6], SATO, S.[7], KITAGAWA, A.[7]
([1]International Christian Univ., [2]RIKEN, [3]Tokyo Univ. of Science, [4]Osaka Univ., [5]Kanazawa Univ., [6]Nihon Univ., [7]National Institute of Radiological Sciences)

A study of ^{57}Fe in-beam Mössbauer spectroscopy using ^{57}Mn provides direct information on exotic state of unstable nuclear probes and on electronic properties of the surrounding materials. Recently, our detection method was successfully improved to obtain in-beam Mössbauer spectra with very large S/N ratio using HIMAC (Heavy Ion Medical Accelerator in Chiba). Here, we demonstrate the validity of this technique applying to a simple metal oxide solid MgO. The ^{57}Mn beam was produced as a nuclear projectile fragment of the ^{5}8Fe beam. The sample used was a MgO polycrystalline disk (50 mm diameter and 5 mm thickness) purchased from Furuuchi Chemical Co. The Mössbauer spectrum of ^{57}Mn implanted into MgO measured at room temperature was relatively simple, which was fitted into the combination of a singlet (δ = -0.91 mm/s) and a doublet (δ = -0.55 mm/s, ΔE_q = 1.3 mm/s). The singlet peak is assigned to high-spin Fe^{2+} surrounded by six O atoms, which substitute Mg^{2+} in MgO crystal. The doublet might be an Fe atom with vacancy of O atoms in neighbor, or an Fe atom in interstitial position of MgO lattice. Density functional calculations with cluster models were performed to make the assignments.

1P01 AMS Analysis of ^{10}Be in the Dome Fuji ice core, Antarctica

IWASAKI, A.[1], SEKINE, T.[2], HORIUCHI, K.[3], MATSUZAKI, H.[4], MURAMATSU, Y.[5], MOTOYAMA, H[6]
([1]Grad. School of Sci., Tohoku Univ., [2]CAHE, Tohoku Univ., [3]Grad. School of Sci. and Engi., Hirosaki Univ., [4]School of Engi., The Univ. of Tokyo, [5]Dep't of Sci., Gakushuin Univ., [6]NIPR)

This study deals with the determination of cosmogenic ^{10}Be in Antarctic ice core samples collected from the Dome Fuji station (at the depth of 290.5-300.5m) to know the secular variation of amounts of the nuclide in the ancient period. Beryllium-10 ($T_{1/2}$ = 1.5 x 10^6 y) is generated via the nuclear spallation reactions of oxygen and nitrogen atoms with galactic cosmic rays.

Ratios of ^{10}Be/^{9}Be in both standards and the samples were measured with AMS at MALT, University of Tokyo, and the annual fluxes of ^{10}Be deposition were estimated by using snow accumulation rates obtained from δ ^{18}O values in this area. It was found that the 10Be flux varied from 20 x 10^4 to 55 x 10^4 atoms/cm^2/y. When the concentration variation of ^{10}Be was compared with the secular distortion of the Cosmogenic nuclide ^{14}C (IntCal04) and Sunspot Number (S.K.Solanki et al. 2005), the similarity was seen in the variation pattern though there was a shift of several decades. The shift at this age suggests the uncertainty of the model of determination of the ice core age.

1P02 Long-term denudation rates of karst surfaces in Japan: estimates from cosmogenic ^{36}Cl in calcite

MATSUSHI, Y.[1], SASA, K.[2], SUEKI, K.[3], TAKAHASHI, T.[2], NAGASHIMA, Y.[2], MATSUKURA, Y.[4]
([1]Micro Analysis Lab., Tandem Accel., The Univ. Tokyo, [2]Univ. Tsukuba Tandem Accel. Complex, [3]RIC, Univ. Tsukuba, [4]Life and Environ. Sci., Univ. Tsukuba)

This study quantified the long-term denudation of bare carbonate rock surfaces by using in-situ-produced cosmogenic ^{36}Cl in calcite. Rock samples were collected from the topmost ~5 cm of exposed pinnacles at several Japanese karst areas under subtropical to subarctic environments. Concentrations of Cl and ^{36}Cl in calcite were determined by accelerator mass spectrometry at UTTAC (University of Tsukuba Tandem Accelerator Complex) with isotope dilution by ^{35}Cl spike. Concentration of Ca and abundances of neutron absorption nuclei were determined by prompt gamma-ray analysis with JRR-3M (Japan Research Reactor No. 3) at JAEA (Japan Atomic Energy Agency) to calculate ^{36}Cl production rate in calcite. The nuclide concentrations were of the order of 10^5 to 10^6 atom g^{-1}. These data were converted to the rate of total denudation that averages chemical and physical processes on the limestone surfaces over 10^4–10^5 yr-timescales. The denudation rates range from ~15 to 60 mm kyr^{-1}, being lowest under temperate climate conditions in the central part of Japan, higher in subarctic northern areas and subtropical southwest areas in Japan. Modeling of this tendency with climate factors at each area indicates a shift in relative importance of chemical (dissolution) and physical (frost shattering) processes in the denudation of karst landforms, depending on mean temperature, annual precipitation, and annual freeze-thaw days. The dissolution rates of carbonate rock surfaces in the southern parts of Japan can be modeled by an Arrhenius-based equation of mean temperature and annual precipitation.

1P03 Measurement of ^{36}Cl in organic substance of soil

AMANO, T.[1], SUEKI, K.[1], TAMARI, M.[1], KITAGAWA, J.[1], SASA, K.[2], NAGASHIMA, Y.[2], TAKAYA, Y.[2], OKI, T.[2], TAKAHASHI, T.[2], KINOSHITA, N.[2], TOSAKI, Y.[2], MATSUSHI, Y.[3], BESSHO, K.[4]
([1]Grad. School of Pure and Applied Sci., Univ. of TSUKUBA, [2]AMS Group, Tandem Accel. Complex, Univ. of Tsukuba, [3]Micro Anal. Labo., Tandem Accel., The Univ. of Tokyo, [4]High Ener. Accel. Res. Organ.)

It is thought that the isotope ratios ^{36}Cl/Cl in soil have several origins such as chlorides, organochlorines, etc. This work is focused on ^{36}Cl/Cl of organochlorines. We used surface soil around Radioisotope center in University of Tsukuba for a test. Chlorides were extracted in the soil with 0.01 mol/l HNO^3, and the extracted solution was called "Extraction sample". The residual soil after extraction burnt in oxygen gas, and the generated gas was trapped with 1.25 mol/l NaOH. It was called "Combustion sample". Additionally, We burnt another soil without extraction and got "Total combustion sample". The AMS measurement results showed that ^{36}Cl/Cl = $(9.8\pm1.0)\times10^{-14}$ of "Combustion sample" was higher than ^{36}Cl/Cl = $(6.0\pm0.5)\times10^{-14}$ of "Extraction sample". The ^{36}Cl/Cl of "Total combustion sample" was $(8.5\pm0.7)\times10^{-14}$ whose value showed between "Combustion sample" and "Extraction sample". By using the core sample soils to 1 m in depth that we got at Rokkasho-mura in Aomori prefecture, we searched the isotope ratios ^{36}Cl/Cl in depth at surface. The isotope ratios ^{36}Cl/Cl of "Extraction samples" and of "Combustion samples" showed a different tendency.

1P04 Improvement of ^{14}C measurement by JAEA-AMS-TONO

Saito-Kokubu, Y.[1], Nishizawa, A.[2], Ohwaki, Y.[2], Nishio, T.[2], Suzuki, M.[1], Ishimaru, T.[1]
([1]Geological Isolation Research and Development Directorate, JAEA, [2]Pesco Corp., Ltd.)

An accelerator mass spectrometer (JAEA-AMS-TONO) installed at the Tono Geoscience has been used to measure ^{14}C concentrations of samples at requests of internal and external laboratories which carry out studies of geoscience. Recently, demands for measurement of small amount of sample are mounting. By request, the AMS and sample preparation technique were improved.

In order to obtain a stable beam, a new temperature control system was equipped with a cesium oven on an ion source. This caused that a variation in temperature of the oven was within 1 °C and stability of the beam enhanced.

For measurement of small amount of sample, the influence of smaller amount of samples than predetermined one and how to fill a cathode with small amount of graphite were studied. On the latter study, two kinds of methods were attempted; (1) iron powder was filled following to fill of the graphite, (2) the graphite and the iron powder were mixed and then the mixture was filled. Counts of ^{14}C in IAEA C1 prepared by the method (1) were gradually decreased and those in the sample of the method (2) were relatively stable. Therefore the method (2) would enable precise isotope ratio measurement.

1P05 Tungsten shield for low level counting

HAMAJIMA Y.
(LLRL, Kanazawa Univ.)

The sintering tungsten shield (diameter: height:

thickness = 130mm : 200mm : 10mm) for low level counting has been made experimentally, set on inside of conventional lead shied, and tested for BKG spectrum of Ge detector in Ogoya Underground Laboratory (OUL), because it is difficult to get low BKG old lead shield. Sum of BKG counting rate from 50 keV to 2000 keV of tungsten shield was 7.9 cpm higher than old lead shield (1.6 cpm), although thickness of tungsten shield is 2/5 of lead shield. Peaks of ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{208}Tl, which were not found in old lead shied, were detected. No peak of ^{210}Pb, environmental neutron induced ^{187}W, and energy region from 70 keV to 200 keV was found. This sintering tungsten shield is not suitable for ultra low level counting in OUL, but may be available for conventional or low level counting in aboveground laboratory.

1P06 Ion-exchange separation for accurate trace analysis of environmental samples for safeguards

MIYAMOTO, Y., YASUDA, K., MAGARA, M., KIMURA, T.
(Nuclear Science and Engineering Directorate, JAEA)

In the isotopic analysis with ICP-MS, tungsten forms polyatomic ions whose molecular weights are 233, 234 and 235, and interferes with the measurement of uranium. Tungsten should be separated chemically from uranium for accurate isotopic analysis of trace uranium. In this work, chemical separation of tungsten from uranium and plutonium in environmental samples for safeguards with a single anion-exchange column was investigated. The mixture of HCl and HF was used as an eluent for anion-exchange separation of tungsten from uranium. It found that 10 ml of the mixture of 8 M HCl and 0.5 M HF was appropriate for the elimination of tungsten from uranium, and 10 - 15 ml of the mixture of 0.1 M HCl and 1 M HF was appropriate for separation of uranium from tungsten. The both anion-exchange separation techniques are able to eliminate almost all tungsten from uranium in environmental samples.

1P07 Particle size distributions of ^{7}Be-aerosols in surface air at Nagano City

MURAMATSU, H., KONUMA, T., MIYAJIMA, A.
(Fac. of Edu., Shinshu Univ.)

The particle size distribution of ^{7}Be-aerosols has been observed using the Andersen-type classifier combined with the high volume air sampler. Aerosol particles were classified and collected on four glass fiber filters, i.e., over 7.0μm in the first-stage, 3.3-7.0μm in the second stage, 2.0 3.3μm in the third stage, and 1.1-2.0μm in the fourth stage, and on a back-up filter for aerosols smaller than 1.1μm in diameter, with a constant flow rate of 566 l/min. About 80% of collected activities were found in the back-up filter, which means that the majority of ^{7}Be activity would attached to aerosols with the diameters less than 1.1μm. Using three data corresponding to particle-size ranges of 3.3-7.0, 2.0-3.3 and 1.1-2.0μm, Activity Median Aerodynamic Diameter (AMAD) was estimated under the assumption that the particle size distribution of ^{7}Be-aerosols in surface air is in a lognormal and unimodal distribution. Estimated AMADs in early summer and in autumn were 0.43μm and 0.55μm, respectively, which should correspond to the mean value of so-called *accumulation mode* aerosols formed by coagulation of ultrafine aerosols.

1P08 Long-term measurement of radioactive nuclides in rainfall and fallen dust

SUZUKI, F.[1], OHASHI, H.[2]
([1]Grad. School of Marine Sci. and Tech., Tokyo Univ. of Marine Sci. and Tech., [2]College of Marine Sci. and Tech. , Tokyo Univ. of Marine Sci. and Tech.)

To investigate the radioactive nuclides transfer from the atmosphere to the ground surface, rainfall and fallen dust samples were collected from 2005 to 2009 with a basin (effective area 0.2m^2). Monthly samples were absorbed to ion exchange resin and measured with a Ge semiconductor detector at ICRR, the University of Tokyo for ^{210}Pb(half-life=22.3y), ^{7}Be (53.3d), ^{40}K (1.27x10^{9}) and ^{137}Cs (30.07y). According to the analysis, remarkable season variations of deposition flux and rain concentration of all the radioactive nuclides were not found. These activity levels were found to depend highly on amount of precipitation.

1P09 Research of Local Fallout ^{236}U produced by Hiroshima Atomic Bomb

KAWAI, K.[1] SAKAGUCHI, A.[2] HOSHI, M.[3] IMANAKA, T.[4] Steier, P.[5] YAMAMOTO, M.[1]
([1]LLRL, Kanazawa Univ., [2]Grad. School Sci., Hiroshima Univ., [3]Res. Inst. Radiat. Bio. and Med., Hiroshima Univ., [4]Res. React. Inst., Kyoto Univ., [5]Wien Univ.)

Since atomic bomb enriched in ^{235}U was exploded over Hiroshima in 1945, the distribution of local fallout has been demanded from a viewpoint of exposure dose evaluation by the residual and fallout radioactivities. The studies on the fallout radionuclides have been mainly made about fission product ^{137}Cs, but fallout distribution has needed further clarification. By this atomic bomb, ^{236}U ($T_{1/2}$ = 2.342×10^{7} y, α-decay) may be expected to be formed by following nuclear reaction; ^{235}U(n, γ). Therefore, present study was made to examine the possibility of ^{236}U detection and possible availability of ^{236}U for clarifying local fallout distribution. Measurements of ^{236}U, ^{137}Cs and $^{239+240}$Pu in surface soils from Hiroshima were performed together with those for soils from Ishikawa Prefecture as background, by using AMS (Wien Univ.) and γ-ray and α-particle spectrometers, respectively. The ^{236}U was clearly detected for all of the soil samples, but these results didn't show any significant difference between ^{236}U/^{239,240}Pu ratios in Hiroshima and Ishikawa surface soils, indicating that the global fallout ^{236}U is dominant contribution and the local fallout ^{236}U produced by Hiroshima atomic bomb is negligibly small contribution.

1P10 Radon concentration in natural water

HORIUCHI, K.
(Otsuma Women's Univ.)

In 1985, Environment Agency specified"Meisui 100 Sen" in all over the country. we have measured the radon concentration in "Meisui 100 Sen" and whose around water for the collection of the basic data about the natural water. Many of "Meisui 100 Sen" is used as a tap water source, daily life water, and tea ceremony water and very familiar with the local people. Most of "Meisui 100 Sen" radon concentration is under 25 Bq/l or so, but a few water which forth out from the granite layer is very high. It is well known that radon concentration and its decay products have given many kinds of valuable information for the study of geochemical phenomena such as flowing mechanism and residence time of spring and groundwater or so.

Recently, people have come to worry about the extension of the decrease and the artificial pollution of the natural water especially “Meisui 100 Sen”.

1P11 Experimental study on radon loss from water sample

YAMADA, N.[1], UESUGI, M.[1], SATO, W.[2], YOKOYAMA, A.[2], NAKANISHI, T.[2]
(Grad. School Nat. Sci. Tech.[1], Kanazawa Univ., Inst. Sci. Eng., Kanazawa Univ.[2])

In the determination of radon in water sample, particular attention should be given to the radon loss from the sample. In a field experiment, radon concentration in running water from a spring decreased with the distance from the welling point. Thereupon, laboratory experiments by using the spring water mentioned above were undertaken. When the spring water was allowed to stand in a beaker in contact with air, radon loss was not observed for 20 minutes. When the spring water was stirred, however, after 20min stirring 20% of radon loss was occurred at 60rpm and 60% loss at 300rpm. The degree of radon loss was roughly proportional to the stirring time at constant stirring speed.

1P12 Uranium series nuclides bound to colloids in borehole groundwater in Kanamaru area, Niigata and Yamagata Prefectures

KANAI, Y., SEKI, Y. and OKUZAWA, K.
(Geological Survey of Japan, AIST)

The behavior in the form of colloid becomes of a great concern recently because the colloidal materials in groundwater may transport the contaminant in the environment. Especially, it is an urgent need to understand detailed behavior of the colloidal materials related to the groundwater pollution and the geological disposal of the high-level radioactive waste (HLW). On the other hand, uranium is a radionuclide existing in nature, and also contained in the HLW, therefore its behavior in nature attracts much attention as a natural analogue. However, such studies are few and have not yet been shown clearly about the uranium series and their colloidal behavior in groundwater. Therefore the behavior of uranium series nuclides and their colloidal behavior in the groundwater in Kanamaru area, Niigata and Yamagata Prefectures, were studied in this study. The uranium series nuclides in the groundwater taken in 2002 at Br1 and Br2 boreholes were analyzed. Uranium content was low in shallow groundwater and high in deep groundwater, and U-234/U-238>1. Th-230/U-234 activity ratio was >1 in shallow groundwater while <1 in deep groundwater. Further measurements conducted in 2005, showed that the excessive Th-230 was not observed. The surface groundwater obtained from Br3-3 borehole (Multi-Packer system) showed Th-230/U-234 >1, and the possibility of the colloid and the suspension was suggested. The water sample taken from the depth of 9.95-11.05m (uranium concentrated layer) of the Br3-3 borehole was preliminary analyzed for the colloid size fractionation analysis.

1P13 Adsorption structure of metal ion on soil including humic acid

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

It is important to determine the concentration of metal ion in natural soil for environmental assessment. But humic acid which is contained in natural soil influences the quantity of adsorbed metal ions on soil. We investigated the quantity of adsorbed metal ion on amorphous clay mineral “allophane” in the condition containing humic acid. We determined concentration of metal elements in humic acid by instrumental neutron activation analysis (INAA). The result shows that calcium is contained most. Therefore, we thought that humic acid has high affinity with calcium and used calcium for this experiment.

We adsorbed calcium on allophane on the condition that the concentration of humic acid was 10ppm and 0ppm in pH 5, 7, 9 and 11. In the system containing humic acid the quantity of adsorbed calcium on allophane increased as pH increased. On the other hand, in the system no humic acid the quantity was almost same from pH 5 to pH 9, but drastically increased at pH 11. Humic acid decreased the quantity of adsorption in an acidic condition and increased that in a basic condition. It is suggested that acid neutralization ability of humic acid and adsorption structure are related to the difference of the quantity of adsorption.

1P14 Fluxes of Pb-210 and Cs-137 in lake sediments

KANAI, Y.
(Geological Survey of Japan, AIST)

Lead-210 and cesium-137 in the lake bottom sediments are used to calculate the sedimentation rate and age of the bottom sediment although each origin and behavior is different. That is, the former flux is considered to be almost constant. On the other hand, the latter flux at present is thought to be the transportation by rolling up of the past deposited nuclide because there is little burst at present. However, the depth profiles of these radionuclides observed in the deposit core don't correspond clearly, and the geological various processes to change in such fluxes are considered. In this study, the depth profile, their fluxes and inventories in the core of the lake bottom sediments (Lake Suwa, Lake Shinji and Lake Nakaumi) were studied in relation to a geological model. The inventories of lead-210 and cesium-137 were almost in linear relationship in the same lake and the surface fluxes of both nuclides also showed a linear relationship. For the flux of lead-210, the atmospheric deposition and the inflow from river are major sources. For cesium-137, the flux supplied from the river might be a major source. The linear correlations observed in the studied lakes suggest that each nuclide did not pile up independently in one lake but both entered in the lake, presumably mixed, homogenized and piled up. This was verified by the model calculation in this study.

1P15 Extremely low background measurement of ^{208}Bi in the soil of the Semipalatinsk nuclear test site in the former USSR.

KOJIMA, S.[1], ARINOBU, T.[1], KOSUDA, C.[1], HAMAJIMA, Y.[2], YAMAMOTO, M.[2], SAITO, T.[3]
([1]Aichi Med. Univ. School Med., [2]LLRL, Kanazawa Univ., [3]Radioisot. Res. Cent., Osaka Univ.)

Residual radioactivities in the surface soil from the Semipalatinsk Nuclear Test Site in the northern part of the Kazakh Republic have been studied. The neutron induced radionuclides, such as ^{60}Co, ^{108m}Ag, and ^{152,154}Eu, and the fission product, ^{137}Cs, were measured in the soil sample collected at the bank of crater which was formed by an underground nuclear detonation on 15 January 1965. The radioactivity of ^{207}Bi in the same sample was measured with a high sensitive well-type Ge detector after radiochemical separation. Although the possible nuclear reactions of ^{207}Bi were offered, the production reaction has not been determined. The object of this study is to explore further investigation about the radionuclide of ^{208}Bi(half life; 3.7x10^5 y) expected to be produced by the similar nuclear reaction. However, the gamma-ray of 2615 keV emitted from ^{208}Bi cannot be measured with a conventional detection system because of the interference by the background gamma-ray from ^{208}Tl. The extremely low background measurement system at Ogoya Underground Laboratory in LLRL, Kanazawa University was adopted to determine the amount of ^{208}Bi in the radiochemically separated Bi sample. The preliminary result was obtained that the count rate of 2615 keV gamma-ray from ^{208}Bi was (2.7±1.5) x10^{-5} cps.

1P16 The coprecipitation behavior of Ba and Ra with manganese dioxide

HINAMI, S.[1], NAKANISHI, T.[2]
([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]Inst. Sci. Eng., Kanazawa Univ.)

The quantitative determination of radium in water sample is usually carried out by the coprecipitation of radium with barium sulfate and by the gamma-ray measurement of the precipitate. However, barium reagents usually contain a certain amount of radium because of the similarity of chemical property between barium and radium; hence the use of barium reagent is limited to the extent that the gamma-ray measurement of low level radioactivity of radium in water sample is not interfered with radium from barium reagent. Accordingly, the removal of radium from barium reagent is important in the measurement of the low level radioactivity of radium in water sample. In this work, the possibility of separation of radium from barium reagent has been investigated using manganese dioxide coprecipitation. The results show that manganese dioxide coprecipitates radium in preference to barium and that the separation of Ra from Ba was improved in the presence of 18-Crown-6 (1, 4, 7, 10, 13, 16-hexaoxacyclooctadecane) in the coprecipitation system.

1P17 Lateral distributions of ^{228}Th activity and ^{228}Ra/^{226}Ra activity ratio of surface water within the Sea of Japan, the East China Sea, and the Sea of Okhotsk

Yoshida, K[1]., Inoue, M[1]., Minakawa, M[2]., Nakano, Y[1]., Kofuji, H[3]., Otosaka, S[4]., Kiyomoto, Y[5]., Shiomoto, A[6]., Hamajima, Y[1]., Yamamoto, M[1].
([1]LLRL, Kanazawa Univ., [2]National Research Institute Fisheries Science, [3]Japan Atomic Energy Agency, [4]Japan Marine Science Foundation, [5]Seikai National Fisheries Research Institute, [6]Tokyo Univ. Agriculture)

The Sea of Japan, one of the largest marginal seas of the Western Pacific Ocean, is surrounded by the Eurasian continent and the Japan Islands, and is connected to the Pacific and other marginal seas by the Tsushima, Tsugaru, and Soya Straits, which have very shallow depths of less than 150m. Therefore, the circulation of the mixed layer water is governed by the complicated lateral movements of the Tsushima Current from the East China Sea and of the Liman Current from the Soya Strait. To study the circulation of water masses and particles within the Sea of Japan, surface water samples were collected from 25 stations within the Sea of Japan, the East China Sea, and the Sea of Okhotsk during the early summer of 2005-2008. By employing low-background γ-spectrometry combined with simple coprecipitation processing on water samples, the activities of ^{226}Ra (half-life $t_{1/2}$ = 1600y), ^{228}Ra (5.75y), and ^{228}Th (1.91y) were determined. It is noteworthy that the ^{228}Th activity of the surface water from the southern areas (the Yamato and Tsushima Basins and the coastal areas along Honshu Island) was lower compared to that of waters from northern areas (the Japan Basin, Yamato Rise, and the coastal areas around Hokkaido). The lateral distribution of ^{228}Th clarifies important aspects of physical movement of geochemical species within the Sea of Japan.

1P18 Distribution of Pu-239,240 concentration in the eastern Pacific

TAKIMOTO, K.[1], SUMI, T.[1], KINOSHITA, N.[2], NAKANISHI, T.[3]
([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]Tandem Accelerator Complex, Univ. Tsukuba, [3]Inst. Sci. Eng., Kanazawa Univ.)

Geographical and depth distribution of anthropogenic radionuclides of Pu-239, 240 ($T_{1/2}$ = 2.411 × 10^4 yr, 6.563 × 10^3 yr) was studied in the eastern Pacific in 2003. As a result of analyses of Pu isotopes in seawater columns and sediment columns, it was found that Pu-239, 240 inventory in water columns was 0.8-5.1 Bq/cm^2 and Pu-239, 240 inventory in sediment columns was 0.3-1.2 Bq/cm^2. The Pu-239, 240 inventory ratio of water-column/sediment-column was calculated to be 3.9-15.2. This inventory ratios compared with those reported for western Pacific gives us information on the sinking behavior of Pu in the eastern Pacific.

1P19 Research on the chemical states of sediments in Tokyo Bay under reducing condition

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

This study was conducted to investigate the chemical states of sediments under reducing condition. We collected core samples of tidal-flat sediments in Tokyo Bay where anoxic zone often appears and in Koajiro Bay by way of comparison. We measured oxidation-reduction potential (ORP) of the sediments on the sampling.

X-ray absorption near edge structure (XANES) spectra of sulfur K-edge of the sediments in estuarine area of Tokyo Bay indicated that two pre-edge peaks which belong to S^{2-}[-II] (2472eV) and SO_3^{2-}[+IV] (2474eV) become larger and that a white peak which belongs to SO_4^{2-}[+VI] (2480eV) becomes smaller as the depth of the samples increases. It means that the oxidation number of sulfur dominantly decreases in a vertical direction as ORP value decreases. In Tokyo bay, anoxic water mass arises and extremely heavy reducing condition is made up on tideland sediments. It is suggested that we can assess the past reducing condition of Tokyo Bay by analyzing the chemical states of elements in the sediments. We also report about the results in Koajiro Bay.

1P20 Distribution of Extractable Organohalogens (EOX) in Tissues and Organs of Great Cormorant (*Phalacrocorax carbo*)

KAWANO, M.[1], SHIKI, Y.[1], KAKOI, T.[1], MATSUDA, M.[1], SUDO, A.[2], MORITA, M.[1]
([1]Faculty of Agriculture, Ehime Univ., [2]Eaglet Office Inc.)

Instrumental neutron activation analysis (INAA) was conducted for the analysis of extractable organohalogens (EOX) in the tissue and organ samples of great cormorant (*Phalacrocorax carbo*) collected in Lake Biwa, Shiga Pref., Japan. The concentration order of EOX was extractable organochlorine (EOCl) > extractable organocbromine (EOBr) > extractable organoiodine (EOI). As the results of investigation into the concentration of EOCl in tissue and organ samples, the highest concentration of EOCl was determined in fat tissue, and the second one was in brain sample, suggesting the presence of different chemical structure compounds in fat tissue and brain of great cormorant from Lake Biwa.

1P21 Determination of trace elements and transformation of proteins in pancreasof zinc-deficient mice

SHIMOYAMA, H.[1], MURAMATSU, W.[1], YAMAMOTO, O.[2], KAWASHIMA, M.[1], SUGANUMA, H.[2], YANAGA, M.[2]
([1]Graduate School of Science, Shizuoka University, [2]Radioscience Research Laboratory, Faculty of Science, Shizuoka University)

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

1P22 Evaluation for Olfactory Transport of Intranasally Administered Insulin-like Growth Factor-I

NAGAOKA, M[1]., WASHIYAMA, K.[2], KOSAKA, K.[3], AMANO, R.[2]
([1]Grad. School Medical Sci., Kanazawa Univ., [2]College Med. Pharmaceutical and Health Sci., Kanazawa Univ., [3]Fac. Med. Kanazawa Univ.)

Insulin-like Growth Factor-I (IGF-I) is thought to be useful for improvement of stroke and neurodegenerative diseases. Although intranasally administered IGF-I is shown to have pathways for direct transport to the mouse brain across blood brain-barrier, the mechanism and interval of time required for transport are still unknown. In the present study, for elucidation of these problems, we studied the temporal IGF-I transport into mouse central nerve system comparatively by administering radioiodine labeled IGF-I solutions intranasally or intravenously.

IGF-I solutions, which were prepared for three types of specific activities, were intranasally and intravenously administered in mouse nasal cavity and in tail vein, respectively. Each group mice were sacrificed under anesthesia and then samples of blood, brain tissues, nasal cavity and skin were excised at some different time point. All samples were measured with γ-counter and calculated uptake rate (%dose/g), except nasal cavity and skin, which were calculated uptake percentage (%dose).

In the group of intranasal administration, the uptake percentage of nasal cavity and uptake rate of blood and brain tissues were increased with a decline in specific activity. Also, uptake rate in olfactory bulb and trigeminal nerve were higher than any other tissues and blood until 60 minutes after administration.

1P23 Production of no-carrier-added Lu-177 capable of labeling antibodies

WATANABE Sa.[1], HASHIMOTO, K.[1], WATANABE, Sh.[1], IIDA, Y.[2], HANAOKA, H.[2], ENDO, K.[2], ISHIOKA, N.[1]
([1]Japan Atomic Energy Agency, [2]Gunma University Graduate School of Medicine)

Production of highly-purified no-carrier-added (nca) ^{177}Lu capable of labeling antibodies was developed using reversed-phase ion-pair liquid chromatography with 2-hydroxyisobutyric acid (2-HIBA) and 1-octanesulfonic acid sodium salt (1-OS) as a complexing and an ion-pairing agent, respectively. ^{177}Lu was produced via the ^{176}Yb (n, γ) ^{177}Yb ($T_{1/2}$=1.911h) → ^{177}Lu process. Enriched $^{176}Yb_2O_3$ targets were irradiated at JAEA JRR-3, dissolved in HCl and loaded on a reversed-phase column to separate the ^{177}Lu from the Yb target. Elution was done with 0.25 M 2-HIBA/0.1 M 1-OS, which has been purified by a cation exchange column and a chelating ion exchange column, respectively, to eliminate metallic impurities such as Ca, Fe and Zn. The pure ^{177}Lu fraction separated was loaded on a cation-exchange column, and then washed with 0.1 M HCl to eliminate 2-HIBA/1-OS. For elimination of further metallic impurities, an anion exchange process was carried out after the cation exchange process of the main separation. The labeling experiment of the antibody with the ^{177}Lu showed that the radiochemical yield of the ^{177}Lu-antibody increased to more than 80% from less than 5% by the elimination of metallic impurities. Consequently, production of nca ^{177}Lu capable of labeling antibodies was succeeded.

1P24 Study of the ^{188}W - ^{188}Re generator using PZC and synthesis of ^{186}Re and ^{188}Re-DMSA

NOGAWA, N.[1], HASHIMOTO, K.[2], KURIHARA, Y.[3], KOIKE, Y.[1], MORIKAWA, N.[1], IJIRI, K.[1]
([1]Radioisotope Center, The University of Tokyo, [2]Japan Atomic Energy Agency, [3]Organisation for the Strategic Coordination of Research and Intellectual Property, Meiji University)

Both ^{186}Re (β ray$_{max}$ 1.08 MeV) and ^{188}Re (β ray$_{max}$ 2.12 MeV) are expected as therapeutic nuclides in the cancer treatment and the pain easing. Poly Zirconium Compound (PZC) has the adsorption capacity of the tungsten tens of times or more the alumina. The basic experiment was done for preparation the small size and practical ^{188}W-^{188}Re generator using PZC. Adsorption yield of ^{188}W to PZC for example showed more than 90% by the heating of 90℃ and 3 hours. Dimercaptosuccinic acid (DMSA) is expected as radioactive medicine for cancer diagnosis and therapy. Synthesis of ^{186}Re and ^{188}Re-DMSA were examined under the alkali condition (pH 14).

1P25 Preliminary evaluation for production and radiochemical separation of an α-emitting radionuclide, ^{230}U

WASHIYAMA, K.[1], TAKAHASHI, N.[2], ARAKI, M.[3], YOKOYAMA, A.[4], SHINOHARA, A.[2], AMANO, R.[1]
([1]College of Medical, Pharmaceutical and Health Sciences, Kanazawa Univ., [2]Grad. Sch. of Sci., Osaka Univ., [3]Grad. Sch. of Natural Sci. and Technol., Kanazawa Univ., [4]College of Sci. and Engineering, Kanazawa Univ.)

Due to short range and high liner energy transfer of alpha particle in human tissue, targeted alpha therapy (TAT) has been applied for cancer therapy and many clinical studies have been performed with the alpha-emitting radioisotopes such as ^{223}Ra, ^{213}Bi, and ^{221}At. However, the very limited availability of ^{225}Ac/^{213}Bi worldwide, and cyclotron capacity made difficult to provide ^{213}Bi and ^{211}At for clinical use. Uranium-230 is a cyclotron produced alpha-emitting radionuclide with a long half-life ($T_{1/2}$ = 20.8d). According to the reaction ^{232}Th (p, 3n) ^{230}Pa and following the β- decay of ^{230}Pa, ^{230}U can be produced within the irradiated target. The daughter of ^{230}U, ^{226}Th ($T_{1/2}$ = 31m) provides a rapid cascade of 4 alpha particles with a 27.7 MeV of total energy. Therefore, ^{230}U is desired to compensate the problem of limited supply of alpha emitter. In this study, we investigated the availability of production of ^{230}U and its radiochemical separation.

Protactinium-230 was produced by irradiation of $^{232}ThO_2$ target on K-70 AVF cyclotron at the Research Center for Nuclear Physics (RCNP), Osaka University. After irradiation, the target was stand for 4 weeks to generate ^{230}U following to the β- decay of ^{230}Pa. The target was dissolved in HNO_3 and converted to HCl solution. The HCl solution was loaded on to an anion exchange column and ^{230}U was separated from the target. After separation, ^{230}U was loaded on another anion exchange column and its daughter, ^{226}Th was eluted. The activity of ^{226}Th and its daughter nuclides were assigned by α spectrometry.

1P26 Estimation of neutron fluxes at the 18 MV medical electron liniac room in Hiroshima University Hospital by an activation foil method and activation activities of the screws of wall sockets.

SAKAMA, M.[1], SAZE, T.[2], OHNO, Y.[3], NAKAMURA, H.[4], TOYODA, A.[4], IIJIMA, K.[4], KOSAKO, K.[5], OISHI, K.[5], MASUMOTO, K.[4]
([1]School Health Sci., Univ. Tokushima, [2]RIC, Univ. Tokushima, [3]Dep. Radiology, Hiroshima Univ. Hospital, [4]KEK, [5]Center Atomic Energy Engin., Insti. Tech., SHIMIZU Corp.)

Recently, from a radiation safety management point of view, it has been discussed to construct guidelines on estimation of neutron fluxes produced via operating various medical small accelerators and to establish uniformly the clearance system related to having neutron activation effects into each accelerator facility. That is, it was the aim of this investigation to accumulate the data on estimation basis of neutron flux measurements at those medical small accelerators by an activation foil method and activation activities of materials in the accelerator facilities. In this work, the neutron fluxes at the 18 MV electron liniac apparatus of Hiroshima University Hospital have been measured by an activation Au foil method and activation activities of screws of wall sockets. For the activation activities of the screws of wall sockets, an 1115.55 keV gamma-ray peak of ^{65}Zn produced via a ^{64}Zn(n, γ) ^{65}Zn reaction was observed at each screw sample in the medical 18MV electron liniac room. The screw sample has contained about 35% zinc and 60% copper which were measured by an X-ray fluorescence analysis, and an isotope abundance of ^{64}Zn is 48.6% in zinc isotopes. As a result of activation activities of ^{65}Zn in the screws of wall sockets, it was also found that the thermal neutron fluxes would be distributed all around the electron liniac room and the measured values are over the range 3.3×10^4 to 9.9×10^4 cm^{-2} s^{-1}.

1P27 Synthesis of water-soluble encapsulated-radioisotope fullerenes

SUEKI, K.[1], NAGASAKI, Y.[1], KAMITSUKASA, T.[1], AKIYAMA, K.[2], TSUKADA, K.[3], ASAI, M.[3], TOYOSHIMA, A.[3], NAGAME, Y.[3]
([1]Grad. School of Pure and Applied Sci., Univ. of Tsukuba, [2]Grad. School of Sci. and Eng., Tokyo Metropolitan Univ., [3]JAEA)

We investigate to trace the synthesis process of water-soluble encapsulated-radioisotope fullerenes using the recoil of nuclear reaction. In order to produce RI@C_{60}, about several ten mg C_{60} fullerenes powder was mixed homogeneously with about several ten mg of oxide (As_2O_3 etc.) and used to the target material. Proton irradiation with beam energy of 13 MeV was performed at the TANDEM in JAEA. The irradiated fullerene samples were dissolved in CS_2 after being filtrated to remove insoluble materials through a membrance filter. The separation of the RI@C_{60} from the solved sample was achieved by two steps HPLC which are the HPLC processes on 5PBB and Buckyprep columns. The yield ratios of monomers labeled with radioisotopes were about 1-2 X 10^{-4}. Toluene solutions of isolated ^{75}Se@C_{60} with C_{60} were vigorously vibrated with saturation KOH aq containing few drops of TBAH (10 % in water) as catalyst at room temperature. The reaction times were 5 - 480 minutes. The resultant brown solution was the passed down a Sephadex G25 size-exclusion gel chromatography column using distilled water as the eluent. We obtained which the activity ratios of ^{75}Se in three phases (Organic phase, KOH aq. phase, and precipitate) as function of reaction time, and which the fraction yields of ^{75}Se in dissolved precipitate through a Sephadex G25 as function time of reaction time. The reaction time was changed the distributions of the activity to a KOH aq. phase and precipitate. As reaction time is long, the activity exists in the KOH phase, and the fraction yields of ^{75}Se@C_{60} increased. The chemical behavior corresponded to the ^{153}Sm@C_{82}.

1P28 Study on HPLC Elution Behavior of Lanthanide Metallofullerenes Using Pyrenyl Stationary Phase

AKIYAMA, K[1]., FURUKAWA, H.[1], SAITO, A.[1], HAMANO, T.[1], SUEKI, K.[2], KATADA, M.[1]
([1]Grad. School of Sci. and Eng., Tokyo Metropolitan Univ., [2]Grad. School of Pure and Applied Sci., Univ. of Tsukuba)

Metallofullerene is one of the most interesting materials because of their characteristic molecular structure and electronic state. However, their properties are not well studied due to the small amount of their production yield which is about 1/1000 compared with that of typical fullerene C_{60}. Radiochromatography is one of the most powerful methods to study the properties of such small amount of materials. In this paper, we report the electric dipole moment of M@C_{82} (M = La, Ce, Pr, Nd, and Gd) estimated from their HPLC retention time determined by the radiochromatography with a pyrenyl stationary phase. The HPLC retention time of M@C_{82} on a pyrenyl stationary phase is determined as 60.2 min for La, 60.9 min for Ce, 61.0 min for Pr, 61.0 min for Nd, and 62.2 min for Gd, respectively. It is considered that logarithm of HPLC retention time is proportional to the square of the electric dipole moment because the separation of metallofullerenes, which have large dipole, on a pyrenyl stationary phase is in terms of the dispersion interaction and the dipole-induced dipole interaction. The evaluated electric dipole moment of M@C$_{82}$ is 5.1 D for Ce, 5.4 D for Pr, 4.5 D for Nd, and 6.3 D for Gd, respectively.

1P29 Extraction behavior of trivalent actinides with HDEHP

TAKAYAMA, R.[1], OOE, K.[1], KOMORI, Y.[1], FUJISAWA, H.[1], KURIYAMA, A.1, KIKUTANI, Y.[1], KIKUNAGA, H.[1], YOSHIMURA, T.[1], TAKAHASHI, N.[1], SAITO, T.[2], MITSUGASHIRA, T.[3], SHINOHARA, A.[1]
([1]Graduate School of Science, Osaka Univ., [2] Radioisotope Research Center, Osaka Univ., [3]Institute for Materials Research, Tohoku Univ.)

The extraction behavior of trivalent actinides into di(2-ethylhexyl)phosphoric acid (HDEHP) has been studied together with a series of lanthanides.

The extraction of Am, Cm, Cf, and lanthanides (excluding Pm) from 0.01 – 1 M nitric acid into 0.01 – 1 M HDEHP in benzene was performed. The distribution ratios of nonradioactive lanthanide isotopes were determined from the concentrations of metals in the aqueous phase and in the organic phase (back extracted to aqueous solution) using ICP-MS, while those of the radioactive actinide and lanthanide isotopes (^{241}Am, ^{243}Cm, ^{249}Cf, ^{139}Ce, and ^{152}Eu) were obtained by γ-ray spectrometry with Ge detectors.

The distribution ratios of Am and Cf increased with decreasing concentration of nitric acid and with increasing concentration of HDEHP, and this behavior was similar to that of lanthanides. However, the distribution ratios of Cm showed different behavior from those of Am and Cf. In this presentation, possible cause of the different behavior observed will be discussed in addition to the systematics of extraction behavior of trivalent actinides.

1P30 Synthesis and Structure of Uranyl (VI) Complex Containing 2-Hydroxyisobutyrate

YOSHIMURA, T., KIKUNAGA, H., SHINOHARA, A.
(Grad. School of Sci. Osaka Univ.)

In this study, we report synthesis and structure of the uranyl (VI) 2-hydroxyisobutyrate complex. The complex was obtained by the reaction of bis (acetato) dioxouranium dihydrate with an excess amount of 2-hydroxyisobutyric acid in water. Uranium (VI) atom is seven-coordinate in a pentagonal-bipyramidal structure. The two oxygen atoms are located at the axial positions with a nearly linear O1-U1-O2 angle. The equatorial positions are coordinated by five oxygen atoms of 2-hydroxyisobutyrate (HIB$^-$) ligands. Two kinds of HIB$^-$ ligands exist in the asymmetric unit. One of the HIB$^-$ ligands links two uranium atoms by the carboxyl group. The other chelates one uranium atom through the hydroxy and carboxyl groups, moreover the carboxyl group bridges the neighboring uranium atom. As a result, a 2D polymeric structure is formed. The IR spectrum showed stretching bands of the carboxyl group of HIB$^-$ at 1614 and 1561 cm^{-1}.

1P31 Cation-exchange behavior of Zr, Hf, and Th in H$_2$SO$_4$/HNO$_3$ mixed solutions – towards to study on sulfate complexation of $_{104}$Rf

Z. J. Li[1], A. TOYOSHIMA[1], K. TSUKADA[1], M. ASAI[1], T. K. SATO[1], T. KIKUCHI[1], N. SATO[1], Y. NAGAME[1]
([1]Advanced Science Research Center, Japan Atomic Energy Agency)

Cation-exchange behavior of Zr, Hf, and Th in H$_2$SO$_4$/HNO$_3$ mixed solutions at [H$^+$] = 1.0 M and [SO$_4^{2-}$] = 4.1 × 10^{-4} M has been studied by a batch method using the carrier-free radiotracers ^{88}Zr, ^{175}Hf, and ^{234}Th. The adsorption sequence on cation-exchange resin is Th >> Hf > Zr. At [H$^+$] = 1.0 M, the distribution coefficients (K_ds) of these elements on the resin gradually decrease with an increase of [HSO$_4^-$], indicating successive formation of sulfate complexes. Further analysis of the slopes at each [HSO$_4^-$] in the log K_d − log [HSO$_4^-$] plot shows that M (SO$_4$)$^{2+}$ and M (SO$_4$)$_2$ (M = Zr, Hf, and Th) are dominant aqueous species in 0.04 − 0.2 M [HSO$_4^-$]. At [SO$_4^{2-}$] = 4.1 × 10^{-4} M, the log K_d values linearly decrease with an increase of log [H$^+$], attributed to the displacement of these elements from the resin by H$^+$. On-line chromatographic behavior of Zr and Hf was also examined with a rapid chemical separation apparatus using the 1.6 mm i.d. × 7.0 mm micro-column. The reaction kinetics of Hf is fast, while the chemical equilibrium of Zr is not achieved. In the symposium, the possibility of study on Rf sulfate complexes will be discussed.

1P32 α-HiB complexation of Zr and Hf as homologues of Rf.

KIKUCHI, T.[1,2], TOYOSHIMA, A.[2], LI, J.[2], TSUKADA, K.[2], ASAI, M.[2], SATO, T.[2], SATO, N.[2], NAGAME, Y.[2], KASAMATSU, Y.[3], FAN, F. L.[4]
([1]Grad. School of Sci. and Eng., Ibaraki Univ., [2]Advanced Sci. Res. Center, JAEA, [3]Nishina Center for Accelerator Based science, RIKEN, [4]Institute of Modern Phys., Chinese Academy of Sci.)

The ion-exchange behavior of Zr and Hf in α-HiB / HNO$_3$ mixed solutions has been studied by a batch method using the carrier-free radiotracers ^{88}Zr and ^{175}Hf. It is found that the α-HiB complex formation of Zr and Hf reaches equilibrium in 180 min and that the ion-exchange behavior of these elements are basically similar to each other. The distribution coefficients (K_d) of Zr and Hf on the cation-exchange resin at [H$^+$]$_{eq}$ = 0.1 M decrease with an increase of [α-HiB $^-$]$_{eq}$, reflecting successive formation of α-HiB complexes.

1P33 Development of liquid scintillation detector for analysis of spent nuclear fuel

KIKUNAGA, H., YOSHIMURA, T., SHINOHARA, A.
(Grad. School of Sci. Osaka Univ.)

We plan to develop an on-line liquid scintillation detector coupled to a capillary electrophoresis apparatus for analysis of spent nuclear fuel. In this work, we have developed a liquid scintillation detector with α / β discrimination and assessed its performances. From the results, it has been found that the liquid scintillation detector has a counting efficiency of 99.6% and a FWHM of 192 keV for the 4784 keV α-peak of ^{226}Ra. Next, a flow measurement system which has two liquid scintillation detectors with U-shaped glass cell connected to each other with 20 cm long of PTFE tube was made. An emulsifier scintillator including ^{241}Am and ^{152}Eu was passed through the detector cell to measure the activities under flow condition and all events were registered in an event-by-event mode in the measurement. Although the flow measurement was successful, peak tailing caused by high viscosity of the emulsifier scintillator was observed.

1P34 Production rate of Pm-146 via proton capture reaction

KINOSHITA, N[1]., MORITA, Y[2]., ARAKI, M[3]., YOKOYAMA, A[4]., TAKAHASHI, N[5].
([1]Tandem Accelerator Complex, Univ. of Tsukuba, [2]Faculty of Sci., Kanazawa Univ., [3]Grad. School of Nat. Sci. Tech., Kanazawa Univ., [4]Inst. of Sci. and Eng., Kanazawa Univ., [5]Grad School of Sci. Osaka Univ.)

P-nuclei, which are heavy nuclei of neutron-deficiency, are considered to be synthesized in supernovae under a temperature of $T>10^9$ K mainly via proton capture and/or neutron emission. The ^{146}Sm nuclide, one of the p-nuclei and also an extinct nuclide, has been known to be abundant in the early solar system based on isotopic anomaly of its daughter nuclide in meteorites. For estimation in nucleosynthesis, spectra of particles and photons as functions of temperature and energy are used to calculate the rates of relevant nuclear reactions. Besides, cross sections of the reactions are also important to estimate the production rate of a particular path for a nuclide. In this work, we focus on the ^{146}Nd (p, n) ^{146}Pm reaction for a path of nucleosynthesis to ^{146}Sm. Cross sections of the reaction have been determined for the energy range of astrophysical importance. For that purpose, proton beam was introduced to the target stack made of isotope-enriched material of ^{146}Nd deposited on highly pure aluminum foils by molecular plating. And the radioactivities from the reaction have been determined by gamma-ray spectrometry. The obtained cross sections are compared with those estimated with the NON-SMOKER and the ALICE calculations. By using the relationship between temperature and production rate, the main path for nucleosynthesis to ^{146}Sm is discussed with the help of the estimate of the NON-SMOKER calculations including those for the other nuclear reactions.

1P35 Excitation Function for Fusion Reaction of the O-16 Projectile with a Deformed Nucleus of Tb-159

KAIYA, H.[1], ARAI, M.[1], ASANO, A.[1], NISHIKAWA, M.[1], OOE, K.[2], KIKUNAGA, H.[2], TAKAHASHI, N.[2], YOKOYAMA, A.[3]
([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]Grad. School Sci., Osaka Univ., [3]Inst. Sci. Eng., Kanazawa Univ.)

In order to study nuclear fusion of deformed nuclei with heavy ions, we focus on lanthanoid targets, which are known to be deformed as actinoids. We have performed an experiment for the synthesis of heavy elements with ^{159}Tb target and ^{16}O projectile. The evaporation residue cross sections were measured by using a stack technique and a gas-jet technique. The cross sections derived from radioactivities of the products were compared with the theoretical values calculated by ALICE code, in which a spherical nucleus is assumed. We compared the experimental values and the theoretical values to discuss parameters influencing the reaction mechanisms.

1P36 Magnetic properties of Co and Fe codoped SnO_2 powders by sol-gel method.

NOMURA, K[1]., OKAMURA, K.[2], YAMADA, K.[2]
([1]School of Engineering The University of Tokyo., [2]Tokyo University of Science.)

SnO_2 powders doped with various Fe^{3+} and Co^{2+} ions were prepared by a sol-gel method. The structure and magnetic properties were characterized by XRD, VSM, and Mössbauer spectroscopy. The rutile types of SnO_2 were determined for all samples. The higher Co concentration, the smaller the lattice is, whereas the higher Fe concentration, the larger the lattice is. The ratio of the lattice constant, c/a, became the smallest for $Sn_{0.98}Co_{0.01}Fe_{0.01}O_2$, which might induce the highest magnetic anisotropy. The hysteresis loops showed the magnetic moment with the maximum 0.121 μB/Co, Fe and with the less coercivity. On the other hand, the coercivity for $Sn_{0.98.5}Co_{0.005}Fe_{0.01}O_2$ showed the large value of 1300kOe. Mössbauer spectra consist of two doublets and a sextet. The intensity of sextet decreased with increasing Co concentration. It suggests that the magnetic interaction of doped Fe ions weakened with Co concentration. It is considered that doublet peak with large QS is due to Fe^{3+} close to oxygen defect, whereas doublet peak with small QS is due to Fe^{3+} substituted to Sn sites.

1P37 Mössbauer study of two-step spin-crossover in Fe (II) complexes of imidazole-containing ligands

IIJIMA, S.[1], HIRATA, Y.[1], YABUKI, S.[1], SATO, T.[2], NISHI, K.[2], MATSUMOTO, N.[2], KAWAMOTO, R.[3], FUJITA, K.[3], MARUYAMA, H.[3], KOJIMA, M.[3]
([1]Natl. Inst. Adv. Ind. Sci. Technol., [2]Res. Inst. Natl. Sci., Kumamoto Univ., [3]Faculty of Sci., Okayama Univ.)

Spin-crossover (SCO) phenomena in some Fe (II) complexes of imidazole-containing ligands were investigated by ^{57}Fe Mössbauer spectroscopy. Complexes [Fe $(H_2L^{2\text{-Me}})_2$] Cl·X showed different temperature profiles of high-spin (HS) molar ratio depending on the size of the counter anion X ($H_2L^{2\text{-Me}}$ = ((2-methylimidazol-4-yl) methylidene) histamine). A two-step SCO transition was observed for the complex with ClO_4^- as X^-, where an intermediate 1/2(LS-Fe^{II} + HS-Fe^{II}) state was detected between ca. 100 and 200 K (LS = low-spin). On the other hand, the complex with PF_6^- exhibited a sharp one-step SCO transition around 200 K. The dinuclear Fe (II) complex $[Fe_2(H_2L')_3](ClO_4)_4$ of the similar ligand L' = imidazole-4-carbaldehyde azine showed LS-Fe^{II} + HS-Fe^{II} state below 180 K, and a sharp one-step SCO transition to LS-Fe^{II} + HS-Fe^{II} state was observed above the temperature. The line-widths of the Mössbauer absorptions and the temperature dependence of the quadrupole splittings suggested that the two Fe sites in $[Fe_2(H_2L')_3](ClO_4)_4$ are not equivalent.

1P38 Syntheses of Mixed Crystals of Assembled Iron Complexes and Their Spin States (II)

DOTE, H[1]., INOUE, K.[1], NAKASHIMA, S.[2]
(Grad. School Sci., Hiroshima Univ.[1] N-BARD, Hiroshima Univ.[2])

We have studied spin-crossover behavior of assembled complexes. In particular, Fe $(NCX)_2(bpp)_2$ (X=S, Se, BH_3; bpp=1,3-bis (4-pyridyl) propane) has very rare structure which is 2D interpenetrated, and Fe $(NCBH_3)_2(bpp)_2$ shows spin-crossover behavior. While Fe $(NCX)_2(bpp)_2$• 2(benzene) has 1D chain structure and shows temperature-independent Fe (II) HS state. In the present study, we performed mixed metals experiment to identify the character of assembled complexes enclathrating benzene molecules.

The structures of complex Fe (Co) $(NCX)_2(bpp)_2$ • 2(benzene) are shown to be similar to those of Fe $(NCX)_2(bpp)_2$ • 2(benzene) by powder and/or single crystal X-ray crystallography. The Mössbauer parameters of Fe (Co) $(NCX)_2(bpp)_2$ • 2(benzene) are similar to those of Fe $(NCX)_2(bpp)_2$ • 2(benzene), suggesting no effect of mixed crystals. And we are now studying the effect of desolvation of benzene molecules in the mixed crystals.

1P39 Preparations and crystal structures for uranyl (VI) coordination compounds with chelate ligands

KITAZAWA, T., KAWASAKI, T.
(Faculty of Science, Toho Univ.)

Actinoid chemistry is highly related to the reprocessing of nuclear fuels and treatment of actioid wastes in the backend chemistry of today's operating nuclear power plant. The fundamental research of the coordination chemistry of uranium complexes allows important information in the field of the backend chemistry. We report herein the synthesis and structures of the new uranyl (VI) coordination compounds with various chelate ligands. The chelate ligands allow 7, 8 and 9 coordination environments.

1P40 Development of real-time RI imaging system for plant nutrient uptake

NAKANISHI, T.M., YAMAWAKI, M., KANNNO, S., ISHIBASHI, H., TANOI, K.
(Grad. School of Agricultural & Life Sci., The Univ. of Tokyo)

It is very important to visualize how nutrients are taken up by the plant. We developed real-time imaging system using conventional beta-ray emitters, ^{32}P, ^{35}S, ^{45}Ca, as well as ^{14}C, except for ^{3}H. The sample was placed in a box where light was irradiated only at an above-ground part of the plant and the roots were kept in dark. Therefore, it was able to prepare cyclic light and dark condition. The beta-rays emitted from the sample were converted to light by a CsI scintilator, which was deposited on FOS (fiber optic plate). After amplification, the light was focused by a lens and was guided to a highly sensitive CCD camera. The image was integrated every 3 minutes, successively. The sensitivity of the imaging was more than ten times higher than that of an imaging plate (IP), with similar resolution. When the chemicals labeled with radioisotopes were applied, it was able to image how these chemicals were taken up by the root as well as translocation and accumulation manner to the above-ground part. In the case of ^{32}P-phosphate, it was found that there was always a preference of translocation to younger tissues in a soybean, rice and other plants.

2S01 Utilization of Radiation – Present and Future Perspective

SHIBATA, T
(J-PARC Center, Japan Atomic Energy Agency)

The radiation and radio-nuclei are wildly used in variety of fields such as research, industrial, agriculture, and medical fields. The economic scale of utilization of radiation and radio-nuclei is the same order of the nuclear power. The future expansion of the radiation usage depends on 1) the extension of neutron- and muon-beam application to wider range of fields, 2) the development of food irradiation to prevent the loss of foods for the future global food problem, 3) development of small neutron sources for usage of neutron beam in the industrial filed, 4) to establish an stable RI supply system in Japan, and 5) an introduction of a reasonable pharmaceutical approval system.

2S02 Nuclear Energy Research and Development -Current Status and Future-

TANAKA, S.
(School of Engineering, The University of Tokyo)

Nuclear energy is evaluated as a sustainable energy source. It is indispensable form the viewpoints of environmental protection and energy sustainability. At present there are 53 nuclear power plants with total capacity of 47.9GWe in Japan. "Framework for Nuclear Energy Policy", which was decided by Atomic Energy Commission in 2005 described three important nuclear policies: ① 30 – 40 % or more of the total electricity production by nuclear even after 2030, ② recycling as the basic nuclear policy, and ③ commercial FBR should be introduced around 2050. For the recycling policy, early and steady operation of Rokkasho Reprocessing Plant is vitally important. Plutonium utilization in LWR is considered as an important first step to effective utilization of nuclear fuel. Disposal of high level radioactive waste is also important to secure nuclear energy utilization. In addition to effective site selection process with public acceptance, fundamental science to improve accuracy of performance assessment is continuously important. For maximum utilization of uranium resources, FBR cycle is most effective. R&D's for demonstration reactor with reprocessing and fuel manufacturing are actively carried out. Considering worldwide increase of nuclear energy in many countries, nonproliferation (safeguards) and nuclear security are becoming more important.

Nuclear chemistry should continue to play an important role in many aspects of nuclear energy research and development.

2S03 Superheavy element chemistry -Frontiers opened by a recoil separator-

HABA, H.
(RIKEN Nishina Center)

Chemical characterization of superheavy elements (SHEs) with atomic numbers $Z \geq 104$ is an extremely interesting and challenging subject in modern nuclear and radiochemistry. Recently, a gas-jet transport system for SHE chemistry has been coupled to the gas-filled recoil ion separator GARIS at the RIKEN Linear Accelerator. We have systematically investigated the performance of the system using ^{206}Fr, ^{245}Fm, ^{255}No, ^{261}Rf, and ^{265}Sg produced in the ^{169}Tm (^{40}Ar, 3*n*), ^{208}Pb (^{40}Ar, 3*n*), ^{238}U (^{22}Ne, 5*n*), ^{248}Cm (^{18}O,5*n*), and ^{248}Cm (^{22}Ne, 5*n*) reactions, respectively. Alpha-particles of these nuclides separated with GARIS and extracted to a chemistry laboratory were successfully identified with the rotating wheel system MANON for α-spectrometry under low background conditions. The high gas-jet efficiencies of over 50% were found to be independent of the beam intensities up to several particle μA. These results demonstrate that the present system is a promising approach for exploring new frontiers in SHE chemistry: (i) the background radioactivities of unwanted reaction products are strongly suppressed, (ii) the intense beam is absent in the gas-jet chamber and hence high gas-jet efficiency is achieved, and (iii) the beam-free condition also allows for investigations of new chemical systems. In the symposium, a chemistry program at RIKEN will be also presented referring to new chemistry apparatuses such as a gas-chromatograph column directly coupled to GARIS and a micro-chemical device for ion exchange and solvent extraction.

2S04 Status and perspectives of muon science at MUSE in J-PARC

NINOMIYA, K1.
([1]Advanced Science Research Center, Japan Atomic Energy Agency)

The new muon science facility (MUSE) at the MLF (Material and Life Science Experimental Facility) in J-PARC (Japan Proton Accelerator Research Complex) has started to supply muon beam for the general users. At MUSE in J-PARC, the 3 GeV 333 μA (1 MW) proton primary beam will be supplied. It is the world's strongest pulsed muon beam source. Two experimental beam ports are now in ready at MUSE (D1 and D2 ports). In the D1-port, the measuring system for muon spin rotation, relaxation, resonance (μSR) method is equipped. Other experiments, such as fundamental studies for elemental analysis by using negative muon, can be performed in the D2-port. In this paper, the status and the new project of muon science at MUSE will be shown.

2S05 The use of synchrotron-based technique for actinide research

IKEDA-OHNO, A.
(Synchrotron Radiation Research Unit (SPring-8), Japan Atomic Energy Agency)

Synchrotron-based analytical methods, such as X-ray absorption spectroscopy (XAS) or X-ray scattering (XS), have recently been recognized as a very powerful tool to investigate the solution / coordination chemistry of chemical substances. These methods have also been applied to the basic- and applied research concerning the actinide species in solution. In this talk, several recent topics related to the actinide speciation study using synchrotrons will be presented, as well as the prospect and future subjects of this research field.

2S06 Application of Mössbauer spectra in the metal complexes

HAYAMI, S.
(Grad. School of Sci. and Tec. Dept. of Chem., Kumamoto Univ.)

The complexes have been attracted attention from the viewpoint of the functionality based on the dynamic electron states. The dynamic electronic states such as the spin states or the oxidation states in the metal complexes develop functions, i.e. spin-crossover, mixed-valence, thermochromism, valence tautomerism, magnetism, conductivity, and so on. In the study for the spin-crossover and mixed valence compounds, iron compounds have been investigated mainly, and ^{57}Fe Mössbauer spectroscopy is convincing measurement means. In this time, The importance of ^{57}Fe Mössbauer spectroscopy for the metal complexes with dynamic electron states and the future development will be discussed.

2S07 Radiochemistry and basic research on radioactive waste disposal and reprocessing

Kirishima, A.
(Institute of Multidisciplinary Research for Advanced Materials, Tohoku University)

Basic researches on the back-end engineering of nuclear fuel cycle including radioactive waste disposal and spent fuel reprocessing are conducted at IMRAM, Tohoku university. In this presentation, three research topics currently conducted are introduced. The first one is "Thermodynamic study on the U complexation with carboxylic acids by micro calorimetry". In this study, ΔG, ΔH and ΔS of U (VI) complexation with carboxylic acids were determined by the calorimetric titration techniques. These data help the deeper understanding of the reaction mechanism. The second topic is "Simplified Modelling of the Complexation of Humic Substance". For the prediction of chemical species distribution in the system containing humic substances, a simplified complexation model is proposed in this study. The proposed model consists of the minimized number of parameters to reproduce the observed complexation behaviour of humic substances. The third topic is "Development of Spent Fuel Reprocessing Process based on Selective Sulfurization". A novel reprocessing process based on selective sulfurization of fission products is proposed. The key concept of this process is utilization of unique chemical property of carbon disulfide, i.e., it works as a reductant for U but works as a sulfurizing agent for minor actinides and fission products. The basic validity of this simple semi-dry reprocessing process having advantages of low radioactive waste volume and risk of the nuclear proliferation is investigated.

2S08 Present Research and Prospects of Environmental Radioactivity

KAKIUCHI, H.
(Dep. of Radioeco. Inst. for Environ. Sci.)

Environmental radioactivity research has been done mainly to clarify the distribution and migration behavior of radionuclides in the environment. Nuclear tests in the atmosphere have been suspended more than 40 years, and the accident of the Chernobyl NPP was passed over 20 years. The moratoriums on unfortunate accidents make it hard to detect its effects now. On the other hand, assessment of an normal release of radionuclides is needed to confirm the safety of residents living near nuclear facilities. We are needed to improve technique for more accurate analysis of radionuclides.

2S09 Radionuclide therapy: A interdisciplinary field of nuclear medicine and radiochemistry

WASHIYAMA, K.
([1]College of Medical, Pharmaceutical and Health Sciences, Kanazawa University)

Radionuclide therapy is one of the fields of nuclear medicine and it uses tumor specific antibody, and/or peptide that labeled with charged-particle (α particle, $\beta -$ particle, Auger electron) emitted radionuclide for treatment of tumors. Beta-emitters, such as ^{89}Sr, ^{90}Y, ^{131}I have been used for clinical field in Japan, and ^{32}P, ^{153}Sm, ^{186}Re have been used in the USA and Europe. These nuclides have suitable half-life and potent $\beta -$ particles. Due to the physical character and attached antibody, yttrium-90 is the first radioimmunotherapeutic agent approved for the treatment of refractory non-Hodgkins lymphoma. Alpha-particles are radiations of high LET. Therefore, the cytotoxicity that induced by α- particles are far more potent than low LET β^{-} particles. Due to the short range of α-particles in tissue, α-emitting radionuclides are suited for targeting micrometastases and single tumor cells. Therefore, astatine-211, ^{213}Bi, ^{223}Ra, and ^{225}Ac have used for clinical trial of leukemia, melanoma, bone metastases, and so on. Other α-emitting radionuclides such as ^{149}Tb, ^{212}Bi, ^{226}Th, ^{227}Th, ^{230}U are also promising for tumor therapy. Especially, radium-223, ^{225}Ac, ^{227}Th, and ^{230}U are the parent of decay chain nuclides. These nuclides act as *in vivo* generator and emit more potent radiation to tumor than single α-, β^--particle emission.

2S10 A study of isotope ratio measurement of ultra-trace nuclear materials in environmental samples

SAITO-KOKUBU, Y.

(JAEA)

Uranium and plutonium of nuclear materials consist of several isotopes. Since their isotopic compositions depend on their origin, their isotope ratios are an effective tool for the identification of the source of uranium and plutonium. In this study, analytical techniques of isotope ratio measurement of ultra-trace nuclear materials were developed and applied to measure isotope ratio of plutonium in sediments and soils collected around Nagasaki city and environmental sample analysis for safeguards.

Ratios of ^{240}Pu/^{239}Pu and concentrations of $^{239+240}$Pu in a sediment core and surface soils collected around the hypocenter of Nagasaki atomic bomb were determined to identify plutonium released from Nagasaki atomic bomb. Depth profiles of the ratio and the concentration in the sediment core indicated depositional record of plutonium released from the atomic bomb for 60 years. The results of ratio of ^{240}Pu/^{239}Pu in the soils allowed us to identify geographical distribution of the plutonium released from the atomic bomb.

JAEA has carried out environmental sample analysis for safeguards. A procedure of a chemical separation and a method for measurement of isotope ratio in particulate nuclear materials by TIMS were developed in order to resolve the problems of analytical technique and supply the analytical demand of actual samples.

2S11 Synchrotron-based X-ray absorption study on actinide complexes in solution

IKEDA-OHNO, A.

(Synchrotron Radiation Research Unit (SPring-8), Japan Atomic Energy Agency)

As compared to typical transition metals, the solution/coordination chemistry of actinides (An) is very unique because of their chemical similarity in the series, as well as their diversity of oxidation states from III to VII. The complex structure of chemical species in solution is the fundamental information to grasp the chemical properties of An in solution. However, conventional analytical technique, such as UV-visible or IR absorption, or NMR spectroscopy, provides no direct information on the structural arrangement of solution species. In this study, the structural identity of An species with different oxidation states has been determined in various aqueous solutions by means of synchrotron-based X-ray absorption spectroscopy. The comparison of the obtained results with other reported data provides us several comprehensive trends of the chemical properties of An in aqueous solution. For instance, tri- and tetravalent An exist as a spherical cation of An^{n+}, while penta- and hexavalent An are forming a transdioxo complex of AnO_2^{m+}. Besides, the highest oxidation state of heptavalent An exists as a negatively-charged square-planar-tetraoxo complex, AnO_4^-. This means that the effective charge of An in solution is completely different from their original charge, and decreased in the following order; An^{4+} (IV) > An^{3+} (III) > AnO_2^{2+} (VI) > AnO_2^+ (V) > AnO_4^- (VII).

2S12 Radiochemical Study on Photonuclear Reaction Mechanism at Intermediate Energies

Sakamoto, K.

(Kanazawa Univ.)

I am greatly honored to receive the Kimura Award from the Japan Society of Nuclear and Radiochemical Sciences. I offer my sincerest gratitude to my mentors during late 1960s, the late Professors, Dr.Paul K.Kuroda and Dr.Sigeo Tanaka who were the former students of Prof. Kimura at the Univ.of Tokyo. My sincere thanks go to the award committee of JSNRS and especially the supporters who nominated me. The work to be presented here has been performed by the collaboration with more than 50 BS,MS and PhD students who passed my lab. and Proffesors, Dr. I. Fujiwara, Dr, S. Shibata, Dr. M. Furukawa and others,with whom the Award be shared.

Our study on photonuclear reactions has been on target nuclei ranging from ^{7}Li to ^{209}Bi at intermediate energies, i. e. bremsstrahlung with the end-point energies of E_0=30-1200 MeV. Irradiation has been performed at the 300MeV-Electron LINAC of Tohoku Univ.,the 1.3 GeV Electron Synchrotron of Institute for Nuclear Study,the Univ. of Tokyo (INS, Now KEK), and partly the 600 MeV Electron LINAC of the Electrotechnical Lab. The study covered the radiochemical yield measurements of photospallation,photopion reactions, photofragmentation, and photofissions of preactinides, ^{197}Au and ^{209}Bi, systematically performed with respect to photon-energy, target mass (A_t) and/or target composition $(N/Z)_t$, product mass (A) and product composition (N/Z). Such a thorough study had not existed in this field, Thousands of the yield data were compiled and found to be smoothly varying functions of these parameters. And the relevant empirical expressions for the reaction yields were derived. The data for the four reaction channels above 140 MeV indicate clear shapes of Δ(1232)-resonance and strong effects of the nuclear medium on the cascade-evaporation process in nuclei with At higher than 110-130 or $(N/Z)_t$ larger than 1.3-1.4. All the results have been updatedly used to test Monte Carlo calculations based on the photon-induced intranuclear cascade and evaporation analysis (PICA) code and its improved versions,and the degrees of the validity of the codes have been demonstrated. New implications for nuclear structure and reaction mechanism have been disscussed by referring to the nuclear models, on which the calculations are based. Kinematic information from additional experiments performed on several targets at the Δ-resonance region has supported the findings from the yield measurements. The results of the yield measurements and the chracteristic features of the four channels are explained separately, though these channels are competitive. Important consequences and perspectives of the work are pointed out relevantly. Some detals are given in J. Nucl. Radiochem, Sci., vol.4, no.2, pp.A9-A31 (2003).

3A01 Prompt Gamma-ray analysis on J-PARC NOBORU

MATSUE H[1]., KASUGAI Y[2]., HARADA M[2]., MAEKAWA F[2]., KUBO K[3]., SAITO T[4].

([1]Quantum Beam Science Directorate, JAEA, [2]J-PARC Center, JAEA, [3]International Christian University, [4]National Museum of Japanese History)

The neutron beam line of NOBORU (BL10) in J-PARC has high flexibility for experimental arrangements. The neutron induced prompt gamma-ray analysis (PGA) system was developed on NOBORU. A germanium detector was used as the gamma-ray spectrometers. The gamma-ray spectrometers of germanium detectors and the data acquisition systems for energy and TOF spectra are prepared. In the experiment, we had PGA data of some materials, such as boron, gold, silver, indium, tantalum etc., and, as trial, we obtained the PGA data of a "Koban", which is a gold coin used in the old days in Japan, and the component fraction of gold and silver in the coin could be analyzed.

3A02 Identification method of ores used for the radiation source in NORM.

FURUTA, E.[1], MINOWA, H.[2], NAKAHARA, H.[3], IWAOKA, K.[4], YONEHARA, H.[4]
([1]Ochanomizu Univ., [2]Jikei Univ. School of Medicine, [3]Tokyo Metropolitan Univ., [4]National Institute of Radiological Sciences)

NORM stands for **N**aturally **O**ccurring **R**adioactive **M**aterial. In this study, NORM means only consumer products. The existence of NORM in our environment raises the radiation exposure dose of the users. This has become a world-wide issue. The sources of NORM are mainly radionuclides in the thorium (Th) and uranium (U) series. However, we have little information about the kinds of ores which are added to NORM. It is well-known that Th and U nuclides usually exist with rare earth elements in ores. So we sought a new method for identifying the kinds of ores added to the NORM from the correlation of the concentration ratios of the Th/U and the Gd/Sm in NORM and ores. The conventional γ-ray spectrometry by a Ge detector for analysis of Th and U and the prompt gamma-ray activation analysis for the determination of Sm and Gd were employed. It was found that the radiation sources of NORM of the consumer products in recent markets were mainly monazite and tourmaline. This method is found to be very useful to identify the ores used for the radiation source in NORM.

3A03 Estimation of the anthropogenic matter ratio of dust particles trapped in snow core

SHOZUGAWA, K.,[1] MATSUO, M.,[1] OSHIMA, M.,[2] TOH, Y.,[2] KIMURA, A.,[2] KOIZUMI, M.[2]
([1]Graduate School of Arts and Sciences, The University of Tokyo, [2]Innovative Nuclear Science Research Group, JAEA)

The anthropogenic matters in air, known as dust particles originated in fuel combustion and refining metals, were trapped in snow during winter season at the mountainous areas. By using the Enrichment Factors (EF) of the elements contained in the dust particles in snow samples, we have evaluated the rate of the anthropogenic matters to soil particles. Analyzing dust particles trapped in snow has a difficulty because of low concentrations of elements and high matrix in insoluble matters. Thus, by applying multiple prompt gamma-ray analysis (MPGA) in addition to INAA, measurements of trace elements which were below the detection limit by conventional methods can be expected.

The snow samples were collected about 1,000g in 2007-2008 winter season at domestic mountainous areas such as Maruyama (Niigata pref.), the peaks of Tateyama (Toyama and Nagano pref.) and Konsei-pass (Tochigi and Gunma pref.). Samples were melted at room temperature, and divided into two aliquots and deposited on membrane filter under a simple clean chamber. γ-γ ray measurements were performed using 24 germanium semiconductor detectors, and measurement for 3,600 sec. was repeated 5 times for every sample. By sorting of each list data, and calculating two-dimensional matrices, the measurement for integration time from 3,600 sec. to 18,000 sec. were obtained. In addition, since the net weight of the dust samples on a filter paper was difficult to measure, EF was calculated from the weight value acquired from comparison with some standard reference materials.

At Murodo, both EF_{Zn} and EF_V showed that measurement for 3,600 sec. had large errors, and sufficient accuracies were not acquired. The results of 10,800 sec. or more were consistent with the results of INAA. Therefore, at least 10,800 sec. for these dust samples were required for measuring time. EF_{Zn} at Murodo indicated about 40, suggesting large effect of anthropogenic activity such as diesel emission from snowplow. The EF_V indicated very lower values than EF_{Zn} at both points; this is explained in terms that particulate matters including vanadium have dissolved into snow. In addition, we will report the ratio of sulfide to sulfate in dust samples, which obtained by XAFS measurements.

3A04 Application of multiple prompt gamma ray analysis (MPGA) to geochemical and cosmochemical samples

WATANABE, R.[1], OURA, Y.[1], EBIHARA, M.[1], OSHIMA, M.[2], TOH, Y.[2], KIMURA, A.[2], KOIZUMI, M.[2], FURUTAKA, K.[2], MURAKAMI, Y.[2]
([1]Grad. School of Sci. and Eng., Tokyo Metropolitan Univ., [2]JAEA)

The multiple prompt gamma-ray analysis (MPGA) is a prompt gamma-ray analysis (PGA) coupled with coincidence counting method using multiple Ge detectors. We studied some conditions for obtaining accurate elemental concentrations in cosmochemical and geochemical samples by MPGA. Reproducibility of counting rates during about 1 day was good and S/N ratio of gamma-ray peaks obtained by sorting using anticoincidence method was about 2 times higher than normal method. We chose 150 – 200 mg of sample as appropriate amount of sample to be analyzed because S/N ratio was increased with an amount of sample up to about 150 mg and S/N ratios are almost same at larger than 150 mg of sample. Determination of Si, Ti, and Fe in Allende and Gold Basin meteorites was tried under suitable conditions obtained in this study, and the concentration values by MPGA were found to be consistent with literature values.

3A05 Determination of trace lanthanoids in ureilite meteorites by radiochemical neutron activation analysis

AIHARA, H., OURA, Y., EBIHARA, M.
(Grad. School of Sci. and Eng., Tokyo Metropolitan Univ.)

Lanthanoids (Rare earth elements; REEs) are a group of elements whose abundances contribute the most significantly in geochemistry and cosmochemistry. In this study, we aimed to determine REEs in several ureilites by radiochemical neutron activation analysis (RNAA). To obtain accurate data of REEs, the contribution of neutron-induced fission reaction of ^{235}U to light REEs (LREEs) was seriously considered and tried to be properly collected.

Ureilite meteorites as well as the Allende meteorite and a peridotite geostandard samples JP-1 were analyzed in this study. These samples were irradiated with neutrons in the JRR-3 reactor of Japan Atomic Energy Agency. The

RNAA procedure was mostly based on that described by Minowa and Ebihara (2003) [1].

Ten REEs (La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb and Lu) were able to be determined. CI Chondrite-normalized REE abundances for ureilites show either so-called V-shaped pattern or LREE-depleted pattern. As ^{140}La, ^{141}Ce and ^{147}Nd are produced by neutron-induced fission reaction on ^{235}U in addition to neutron-capture reaction of individual target nuclides (^{139}La, ^{140}Ce and ^{146}Nd, respectively), their contributions were estimated and corrected for. As JP-1 has high U/LREEs ratios, collections reach 4%, 21% and 25% for La, Ce and Nd, respectively.

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

3A06 Activity ratio of ^{230}Th/^{232}Th in the products from Asama volcano, Japan

KURIHARA, Y.[1], Takahashi, M.[2], SATO, J.[3]
([1]Org. Strategic Coordinat. Res. Intellect. Prop., Meiji Univ., [2]Cent. Res. Inst. Electr. Power Industry, [3]Meiji Univ.)

The generation of magmas at the subduction zone is generally inferred to be induced by the partial melting of the mantle wedge by the addition of fluid derived from the subducting plate. Observations were carried out on the radioactive disequilibria between ^{238}U and ^{230}Th in the products (4th C, 1108 and 1783 eruptions) from Asama volcano, Japan. Measurement of activity concentrations and activity ratios of U and Th isotopes in the samples was made by isotope dilution analysis coupled with alpha-ray spectrometry. Activity ratios of ^{238}U/^{230}Th in the products were greater than unity, being enriched in ^{238}U relative to ^{230}Th, which was often observed for fresh products from subduction zone volcanoes. The ^{230}Th/^{232}Th activity ratio in the products from Asama volcano and from Izu-Mariana arc volcanoes ranged from 0.91 to 0.95 and from 1.02 to 1.67, respectively, and the ^{238}U/^{232}Th activity ratio ranged from 0.98 to 1.16 and from 1.09 to 2.81, respectively. The (^{230}Th/^{232}Th)-(^{238}U/^{232}Th) data set for Asama volcano formed a cluster on the diagram, while those of Izu-Mariana arc volcanoes formed another cluster apparently apart from each other, suggesting that the ^{230}Th/^{232}Th (= ^{238}U/^{232}Th) activity ratio in the magma source may possibly be different between these volcanoes.

3A07 Study on dynamics and sources of POC in river systems using ^{14}C as a tracer

NAGAO, S[1]., ARAMAKI, T.[2], IRINO, T.[3], UCHIDA, M.[2], SHIBATA, Y[2]., TOGAWA, O.[4]
([1]INET, Kanazawa Univ., [2]Enviorn. Chem. Divi., NIES, [3]Fac. of Environ. Earth Sci., Hokkaido Univ., [4]Nucl. Sci. Eng. Direct., Japan Atomic Energy Agency)

In order to understand transport behavior of particulate organic matter (POM) in river systems, we studied characteristics of POM in seven rivers with different watershed conditions. River water samples were collected at a station in lower region each river. The suspended solids (SS) were concentrated from river waters using a single-bowl continuous flow centrifugation. The SS samples were measured for organic carbon content, Δ^{14}C and δ^{13}C values. The Δ^{14}C of POC ranges from −296 to +20‰. On the basis of Δ^{14}C-δ^{13}C plots, carbon isotopic signatures can be divided into three groups. These results indicate that Δ^{14}C and δ^{13}C values of POC depend on watershed conditions and sources of riverine POC.

3A08 Separation of dissolved ^{85}Kr in groundwater

Ohta, T.[1], Mahara, Y.[1], Momoshima, N.[2], Inoue, F.[3], Ikawa, R.[4], Shimada, J.[5] Kubota, T.[1], Taniguchi, M.[6]
([1]RRI, Kyoto Univ., [2]RIC, Kyushu Univ., [3]Dep. Chem., Fac. Sci., Kyushu Univ., [4]AIST, [5]Fac. Sci., Kumamoto Univ., [6]RIHN)

We investigated extraction of the ^{85}Kr from groundwater having an ultra-low concentration with a Kr-extraction system using a hollow fiber membrane. Our aim was to develop a system able to effectively extract Kr isotopes from a large volume of groundwater (10^4 L) in field studies for measurement of ^{85}Kr radioactivity. Before testing our Kr-extraction system in the field, we investigated the efficiency of Kr extraction from water by extracting trace amounts of Kr in the laboratory. This extraction method can be applied to 10^4 L of water, though a flow velocity at 24 L/min is preferable. We then used our method to extract ^{85}Kr from groundwater at field, Japan. We showed here that Kr isotopes, which reflect residence time of groundwater, can easily be obtained by the system.

3A09 Sequential ion-exchange separation and analysis of trace uranium, thorium, lead and lanthanides in environmental samples

MIYAMOTO, Y., YASUDA, K., MAGARA, M., KIMURA, T.
(Nuclear Science and Engineering Directorate, JAEA)

Abundance and isotopic ratios of trace U, Th, Pb and lanthanides in a sample play a key role to investigate features of the sample. From these ratios of the key elements, information on origin and history of the sample can be obtained. The key elements should be separated from the co-existing and interfering elements in a sample to analyze accurately the abundance and isotopic ratios of the key elements by ICP-MS. In this work, nano grams of U, Th, Pb and lanthanides in a geological reference rock were sequentially separated with a single anion-exchange column. Mixture of CH_3COOH, HCl and HNO_3 were used as the eluents for this separation. For the Th separation, 22 ml of the mixture of 70% CH_3COOH and 1 M HCl was effective. By addition of 0.005 M HF to the eluents, the recoveries of Th and U were improved. Alkaline metals, alkaline earth metals and Fe were effectively eliminated from the key elements. By using this technique, it is possible to simplify the complicated traditional processes without any special ligands, columns and equipments.

3A10 Distribution of ^{241}Am concentration in water column of eastern Pacific in 2003

NAGAOKA, M.[1], SUMI, T.[1], KINOSHITA, N.[2], YOKOYAMA, A.[3], NAKANISHI, T.[3]
([1]Grad. School of Nat. Sci. Tech., Kanazawa Univ., [2]Tandem Accelerator Complex, Univ. Tukuba, [3]Inst. of Sci. Eng., Kanazawa Univ.)

Americium-241 ($T_{1/2}$ = 432.2 yr) profiles have been determined in the eastern Pacific in 2003. The vertical distribution of ^{241}Am showed similar pattern to those of ^{239,240}Pu ($T_{1/2} = 2.41\times10^4$ yr, 6.56×10^3 yr). The activity ratios of ^{241}Am / ^{239,240}Pu were low in surface and bottom layer. It suggests that ^{241}Am is scavenged from surface more rapidly than ^{239,240}Pu and ^{239,240}Pu is redissolving from sediment..

3A11 Determination of ^{226}Ra by liquid scintillation spectrometry using ^{223}Ra as yield tracer

UESUGI, M.[1], HINAMI, S.[1], YOKOYAMA, A.[2], NAKANISHI, T.[2]
([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]Inst. Sci. Eng., Kanazawa Univ.)

Drinking water or groundwater contains very small amount of ^{226}Ra. The preconcentration of radium is essential for the analysis of the nuclide in water sample. It is well known that barium sulfate coprecipitates radium quantitatively, but the amount of barium is limited in applying liquid scintillation spectrometry or α-ray spectrometry to count alpha-radioactivity of ^{226}Ra. In this work, the preconcentration of Ra by hydroxyl apatite coprecipitation, separation of Ra from Ca, and liquid scintillation spectrometry were examined by using ^{223}Ra as a yield tracer. The results are as follows: (1) 30 mg/L of Ca^{2+} carrier can carry down more than 95% of radium into hydroxyl apatite, (2) by adding citrate, radium was effectively separated from calcium by using cation exchange resin column, (3) by transferring cation exchange resin which sorbed Ra into liquid scintillation vial and by adding liquid scintillator, ^{222}Rn grown from ^{226}Ra and extracted into scintillator was effectively counted by liquid scintillation spectrometer.

3A12 Estimation of soil-to-plant transfer of radium-226

TAGAMI, K., UCHIDA, S.
(National Institute of Radiological Sciences)

Among the environmental transfer parameters that have been used in the mathematical models for radiation dose assessment for humans, soil-to-plant transfer factor (TF) is of importance. The parameter is calculated as the plant/soil concentration ratio; however, for ^{226}Ra, the number of reported TFs is still limited due to the low concentration in plant samples. In this study, a statistical approach was used to estimate TF-Ra instead of direct measurement of the radionuclide in plants and soils. The results showed high correlations between ^{226}Ra and U concentrations in soils (r=0.816, $p<0.001$), and between ^{226}Ra and Ba concentrations in plants (r=0.887, $p<0.001$) in the Student's t-test. Using these results, TF-Ra could be estimated with good accuracy. Indeed, the difference between estimated and measured TF-Ra was within a factor of 2 for each crop species. The method could estimate TF-Ra under the soil-to-plant systems where no significant enhancements of U (^{226}Ra) and/or Ba concentrations in soil are expected. The proposed empirical method could serve as a screen for potential exposure to ^{226}Ra from plant ingestion by humans.

3A13 ^{210}Pb deposition in the Far East Asia

HIROSE, K.[1], KIKAWADA, K.[1], DOI, T.[2], SU, C.-C.[3], YAMAMOTO, M.[4]
([1]Faculty of Sci. Technol., Sofia Univ., [2]NIES, [3]National Taiwan Univ., [4]LLRL, Kanazawa Univ.)

Our aim of this study is to have better understanding of spatial and temporal variations of the ^{210}Pb deposition in the Far East Asia. To achieve our purpose, a comprehensive data set of monthly ^{210}Pb deposition, which includes several time-series data and spatial distribution data in 13 stations of Japan and 2 stations in Taiwan, was analyzed. The ^{210}Pb deposition in the Far East Asia exhibited a latitudinal distribution. Especially, the Japan Sea side stations were characterized by anomalous high ^{210}Pb deposition as pointed out by Yamamoto et al., (2006). The ^{210}Pb depositions in the Far East Asia showed seasonal change with high in winter having large geographical variability and low in summer having small geographical variability. We examine ^{210}Pb deposition behaviors in winter and summer, respectively. The results revealed that the relationship between the ^{210}Pb deposition and precipitation amounts in the Japan Sea side stations in winter differs from that in other sites whereas there is no difference of the ^{210}Pb deposition behavior between sampling sites in summer. The ^{210}Pb concentrations in rainwater in winter showed inter-annual variability. This may be related to climatological factors such as ENSO.

3B01 Production of Radioisotopes for Nuclear medicine using fast neutron reactions

HATSUKAWA, Y., HASHIMOTO, M., NAGAI, Y., KIN, T., SEGAWA, M., HARADA, H., KONNO, C., OCHIAI, K., TAKAKURA, K.
(Japan Atomic Energy Agency)

^{99m}Tc is the most important radioisotope used as nuclear medicine. The present world demand for ^{99}Mo is about 450 TBq/week which is mostly produced by using only five nuclear reactors in Canada, Belgium, France, Netherlands and South Africa. Recently, two of the present authors proposed a new route to produce ^{99}Mo by the ^{100}Mo $(n, 2n)$ ^{99}Mo reaction, which has the following characteristic features.[1)] First, the reaction cross section is large, about 1.5 barn at 12<En<17 MeV, which is 10 times larger than the thermal-neutron capture cross section of ^{98}Mo. Second, the cross sections of the (n, α), (n, np), and (n, p) reactions are less than a few mb at En = 14 MeV. Third, a large amount of ^{100}Mo target materials can be used, compared to that for proton beam irradiation on ^{100}Mo. Fourth, intense neutrons with energy of 12-17MeV are already available. In the present work we have measured all γ-rays emitted from activities produced by bombarding a natural Mo target with neutrons from the D (^{3}H, n) ^{4}He reaction at Fusion Neutronics Source Facility (FNS) at Japan Atomic Energy Agency (JAEA) to study characteristic features mentioned above more in detail. The neutron flux was about 10^{12} n/cm^{2}s. The experimental results at FNS will be discussed in the conference.

Reference

1) Y. Nagai and Y. Hatsukawa, J.Phys. Soc. Jpn 78, 033201 (2009)

3B02 Production of rare earth multitracer from ^{nat}Hf target

Y. EZAKI, H. HABA, Y. KASAMATSU
(RIKEN Nishina Center)

A multitracer including many radioisotopes of rare earth elements (REEs) is very useful to simultaneously trace the behavior of REEs under an identical experimental condition. In this paper, we propose a novel method to produce the REE multitracer using ^{nat}Hf as a target.

The ^{nat}Hf foil of 165 mg/cm^2 in thickness was irradiated with a 135 MeV/nucl. ^{14}N beam from the K540 RIKEN Ring Cyclotron (RRC). The beam intensity was about 0.6 particle μA. For effective productions of short- and long-lived radionuclides, two separate ^{nat}Hf targets were irradiated for 0.5 h and 59 h, respectively. After the irradiations, the REEs were chemically separated through coprecipitations with calcium fluoride and iron hydroxide followed by anion- and cation-exchange methods. The REE multitracers were then subjected to γ-ray spectrometry using a Ge detector.

As a preliminary result, about 20 radionuclides of Y, Ce, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu were identified in the short irradiation. Chemical yields of over 80% were obtained for those nuclides. A separation factor for the Hf target material was evaluated to be 3×10^{-3} based on the 343-keV γ peak of ^{175}Hf. In the conference, the results on the long irradiation will be also presented together with those on the conventional REE multitracer from a ^{197}Au target.

3B03 Measurement of Pa-231 photofission cross-section by fission track method

NISHIKAWA, M.[1], OHTUKI, T.[2], HIROSE, K.[2], SATO, W.[3], YOKOYAMA, A.[3], NAKANISHI, T.[3]
([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]Lab. Nucl. Sci., Tohoku Univ., [3]Inst. Sci. Eng., Kanazawa Univ.)

Fission track method has been applied to the photofission reaction of ^{231}Pa. Quartz and muscovite were used to detect fission fragments. Natural uranium (^{n}U) and ^{232}Th were used as reference nuclides. Each nuclide was sandwiched between quartz and muscovite to irradiate with bremsstrahlug at Tohoku Univ. After the irradiation, quartz and muscovite detectors were subjected to chemical etching. As for the etching condition, etching for 15 minutes by using 28% solution of ammonium hydrogen fluoride proved suitable for quartz, and etching for 40 minutes by using 48% hydrogen fluoride proved suitable for muscovite. The 'number of fission tracks/number of target nuclides' ratios were compared between simultaneously irradiated ^{231}Pa, ^{n}U, and ^{232}Th to evaluate the photofission cross-section of ^{231}Pa.

3B04 Cross section measurements of neutron-induced reactions for Cu and Nb in the intermediately energy region (II)

OMOTO, T.[1], NINOMIYA, K.[1, 2], NAKAGAKI, R.[1], TAKAHASHI, N.[1], SEKIMOTO, S.[3], UTSUNOMIYA, T.[3], YASHIMA, H.[3], SHIBATA, S.[3], SHIMA, T.[4], KINOSHITA, N.[5], MATSUMURA, H.[5], HAGIWARA, M.[5], IWAMOTO, Y.[2], SATOH, D.[2], M. W. CAFFEE[6], IMAMURA. M.[7], NISHIIZUMI, K.[8], SHINOHARA, A.[1]
([1]Grad. School Sci., Osaka Univ., [2]JAEA, [3]Res. Reactor Inst., Kyoto Univ., [4]RCNP, Osaka Univ., [5]KEK, [6]Dep. Phys., Purdue Univ., [7]National Museum of Japan History, [8]Space Sci. Lab., California Univ.)

Neutron induced nuclear reactions in the intermediate and high energy region (>50 MeV) are important to investigate whole energy dependence of hadron induced nuclear reactions. It is because there are few experimental studies of neutron induced reactions above 50 MeV. There are large deviations on the nuclear reactions between neutron and proton induced reactions in the low energy region due to the charge difference. In this study, monoenergetic 282 MeV neutron flux was obtained from the two proton irradiations for ^{7}Li target with 300 MeV at the N0 course in the Research Center for Nuclear Physics, Osaka University. We performed the monoenergeic neutron irradiations for cupper and niobium metals. Production cross sections of the radionuclides were determined with a gamma-ray counting method by using high purity germanium detectors. In this presentation, we discuss the energy dependence of cross sections by comparing 282 MeV and 368 MeV (our previous work) neutron induced reactions, and examine the nuclear reaction mechanism through comparison between the cross sections of neutron and proton induced reactions.

3B05 Measurement of the half-life of Th-229g

KIKUNAGA, H.[1], MITSUGASHIRA, T.[2], SUZUKI, T.[3], NOMURA, M.[3], SHINOHARA, A.[1]
([1]Grad. School of Sci., Osaka Univ., [2]IMR, Tohoku Univ., RLNR, [3]Tokyo Inst. Technol.)

The half-life of ^{229g}Th has been reevaluated. Uranium isotopes which were determined the isotopic ratio with TIMS were purified with anion exchange columns to eliminate thorium isotopes. In order to determine the isotopic ratio of ^{232}U to ^{233}U, a part of the uranium isotopes was subjected to α-spectrometry. The purified uranium isotopes were left to stand for 3-289 d to allow the growth of thorium isotopes. The thorium isotopes were separated from the uranium isotopes and subjected to α-spectrometry to determine the isotopic ratio of ^{228}Th/^{229}Th. From those isotopic ratios and the half-life of ^{228}Th ($T_{1/2} = 1.9116\pm0.0016$ y), ^{232}U ($T_{1/2} = 68.9\pm0.4$ y), and ^{233}U ($T_{1/2} = 159200\pm200$ y), we estimate the half-life of ^{229}Th to be 7896 ± 66 y.

3B06 Nuclear deformation effect on the excitation function of heavy ion fusion reaction on rare earth targets with the O-16 projectile

Asano, A.[1], Kaiya, H.[1], Araki, M.[1], Nishikawa, M.[1], Ooe, K.[2], Kikunaga, H.[2], Takahashi, N.[2],
Yokoyama, A.[3],
([1]Grad. School Nat. Sci. Tech., Kanazawa Univ., [2]Grad. School Sci., Osaka Univ., [3]Inst. Sci. Eng., Kanazawa Univ.)

We have performed an experiment for the synthesis of heavy elements with oxygen-16 and Tm-169 to compare the results with Ba and La targets. The Tm target is more deformed than the others. The formation cross sections of evaporation residues through the nuclear fusion process were measured by using a stack technique and a gas-jet technique. The cross sections were derived from radioactivities of the products. The obtained excitation functions of the La+O and the Tm+O systems are compared with each other to discuss influence of nuclear deformation.

3B07 Study of sub-barrier fusion hindrance in ^{19}F + ^{209}Bi reaction

NISHINAKA, I.[1], KASAMATSU, Y.[2], TANIKAWA, M.[3], GOTO, S.[4], ASAI, M.[1]
([1]Advanced Sci. Res. Center, JAEA, [2]Nishina Center, RIKEN, [3]Fac. Sci. , Univ. Tokyo, [4]Fac. Sci. , Niigata Univ.)

The fusion-fission cross sections for ^{19}F + ^{209}Bi at sub-barrier energies were determined by a radiochemical method in order to study sub-barrier fusion hindrance in heavy-mass systems. The irradiations of the ^{209}Bi target with 83-135 MeV ^{19}F ions were carried out at the JAEA tandem accelerator. After irradiation, the target, the backing and the catcher foil in which fission fragments were collected were dissolved in 6M HCl solution. The radiochemical separations of molybdenum (VI) from fission product mixture were carried out by using ion-exchange techniques. Precipitation of Mo with α-benzoin-oxime was applied to the preparation of samples for γ-ray spectrometry. Fission cross sections were obtained from the cross sections of ^{99}Mo by using the fractional yield of ^{99}Mo to the total fission yield which was deduced from the mass yield curves. The fusion-fission excitation function for ^{19}F + ^{209}Bi at sub-barrier energies was determined down to nearly two order magnitude smaller than the data measured in the energy range of 88-123 MeV by silicon surface barrier detectors. We will discuss the sub-barrier fusion hindrance in the present reaction, comparing the theoretical calculations and the systematics.

3B08 Development of a new gas-filled recoil ion separator GARIS-II

KAJI, D.[1], MORIMOTO, K.[1], SATO, N.[2], YONEDA, A.[1], MORITA, K.[1]
([1]Nishina Center for Acc. Based Sci., RIKEN, [2]Advanced Sci. Research Center, JAEA)

We designed a new gas-filled recoil ion separator GARIS-II. A gas-jet transport system coupled to GARIS-II as a pre-separator will be a promising tool for next-generation superheavy element chemistry, i.e., identifying SHE nuclides under low background conditions with high efficiency of the gas-jet transport. GARIS-II consists of five magnets arranged in a Q_v-D-Q_h-Q_v-D configuration. This configuration is the first design for the purpose of SHE study. Ion optical characteristics were analyzed using the computer code TRANSPORT. The solid angle is increased from 12.2 to 20.2 msr, approximately 1.7 times higher than GARIS. The total path length of the separator is 5.12 m. GARIS-II will be able to use various filled gas, such as He and He/H_2 mixture. The separator will be able to transport the recoil ion with magnetic rigidity $B\rho$ =2.44 Tm.

3B09 Production of ^{261}Rf for chemical studies using the gas-jet transport system coupled to GARIS

HABA, H.[1], OOE, K.[2], OZEKI, K.[1], KASAMATSU, Y.[1], KAJI, D.[1], KUDOU, Y.[1], KOMORI, Y.[2], SATO, N.[3], SHINOHARA, A.[2], MORITA, K.[1], MORIMOTO, K.[1], YONEDA, A.[1]
([1]RIKEN, [2]Osaka Univ., [3]JAEA)

A gas-jet transport system for superheavy element chemistry has been coupled to the gas-filled recoil ion separator GARIS at the RIKEN linear accelerator. In this work, we have investigated the performance of the system using ^{261}Rf produced in the ^{248}Cm (^{18}O, *5n*) ^{261}Rf reaction. Alpha-particles of ^{261}Rf separated with GARIS and extracted to a chemistry laboratory were successfully identified with a rotating wheel system for α-spectrometry under extremely low background conditions. By comparing with the spectrum measured with a focal plane Si detector in a separate experiment, the gas-jet transport efficiency of ^{261}Rf was evaluated to be 52±12%. The transport efficiency of GARIS was 7.8±1.7% for the focal plane of 100-mm diameter, referring to the cross section of 13 nb measured at JAEA. The present result demonstrates that the GARIS/gas-jet system is a promising approach to explore new frontiers in SHE chemistry: (i) the background radioactivities originating from unwanted reaction products are strongly suppressed, (ii) the intense primary heavy-ion beam is absent in the gas-jet chamber, and hence high gas-jet transport efficiency is achieved, and (iii) the beam-free conditions also make it possible to investigate new chemical systems that were not accessible before.

3B10 Production of long-lived Fm and Es tracers using a Cf target and its application to nuclear spectroscopy

ASAI, M.[1], TSUKADA, K.[1], KASAMATSU, Y.[1], SATO, T.K.[1], TOYOSHIMA, A.[1], NAGAME, Y.[1]
([1]Adv. Sci. Res. Center, JAEA, [2]Nishina Center, RIKEN)

We have successfully obtained a large amount of long-lived Fm and Es tracers which can be used in various kinds of heavy-actinide chemistry experiments. Fm and Es isotopes were produced with a Cf target and a ^{12}C beam from the JAEA tandem accelerator. Reaction products recoiling out of the target were collected with a gas-jet transport technique. After 3-day irradiation, Fm and Es isotopes were chemically separated from the collected products. By measuring α- and γ-energy spectra, we evaluated an amount of each Fm and Es isotope, and other radioactive impurities. We also performed α-γ coincidence measurements for ^{253}Fm using the same source.

3B11 Productions and decay properties of a new isotope of ^{263}Hs

KAJI, D.[1], MORIMOTO, K.[1], SATO, N.[2], HABA, H.[1], ICHIKAWA, T.[3], IDEGUCHI, E.[4], KOURA, H.[2], KUDO, Y.[1], OZAWA, A.[5], OZEKI, K.[1], SUMITA, T.[1,6], YAMAGUCHI, T.[7] YONEDA, A.[1], YOSHIDA, A.[1], MORITA, K.[1]
([1]Nishina Center for Acc. Based Sci., RIKEN, [2]Advanced Sci. Research Center, JAEA, [3]Yukawa institute for theoretical physics, Kyoto Univ., [4]Center for Nucl. Study, Univ. of Tokyo, [5]Univ. of Tsukuba, [6]Tokyo Univ. of Science, [7]Saitama Univ.)

A new neutron deficient hassium (Z=108) isotope of ^{263}Hs was identified via two different reactions of ^{206}Pb (^{58}Fe, n) and ^{208}Pb (^{56}Fe, n) by using the gas-filled recoil separator GARIS. During irradiation of ^{206}Pb with the ^{58}Fe beam and irradiation of ^{208}Pb with the ^{56}Fe beam, 8 decay chains and 1 decay chain, respectively, were observed. The properties of decay events obtained by irradiation of ^{58}Fe on ^{206}Pb match well with those by irradiation of ^{56}Fe on ^{208}Pb. The mean α-decay energies for the 3 groups are 10.82, 10.55, and 10.37 MeV, respectively. The half-life of ^{263}Hs is determined to be 0.60 ms.

3B12 Productions and decay properties of ^{264,265}Hs

SATO, N.[1], KAJI, D.[2], MORIMOTO, K.[2], HABA, H.[2], ICHIKAWA, T.[3], IDEGUCHI, E.[4], KOURA, H.[1], KUDOU, Y.[2], OZAWA, A.[5], OZEKI, K.[2], SUMITA, T.[2,5], YAMAGUCHI, T.[6], YONEDA, A.[2], YOSHIDA, A.[2], MORITA, K.[2]
([1]Advanced Science Research Center, Japan Atomic Energy Agency, [2]Nishina Center for Accelerator Based Science, RIKEN, [3]Yukawa Institute for Theoretical Physics, [4]Center for Nuclear Study, Univ. of Tokyo, [5]Univ. of Tsukuba, [6]Tokyo Univ. of Science, [7]Saitama Univ.)

The deformed proton shell gap at Z = 108 and the neutron shell gaps at N = 152 and 162 have been found from the systematic studies of α-spectrometry of the heaviest nuclei. The doubly deformed magic nucleus ^{270}Hs (Z = 108, N = 162) and its neighbor nuclei had been studied in detail. However, the properties of neutron-deficient Hs nuclei are not well known.

In this work, the production and decay properties of ^{264,265}Hs produced in the ^{208}Pb (^{58}Fe, *xn*) [x = 1,2] and ^{207}Pb (^{58}Fe, *n*) reactions were investigated. The experiment was performed at the RILAC facility in RIKEN. The evaporation residues (ERs) were separated from the primary beam with the gas-filled recoil ion separator, GARIS. In the focal plane of GARIS the products were implanted into a position-sensitive Si detector for measuring the arrival of ERs and subsequent α-decays or spontaneous fission from ERs.

We have measured production cross-sections of ^{264,265}Hs in the ^{58}Fe + ^{208}Pb reaction at several bombarding energies. For the ^{208}Pb (^{58}Fe, *n*) ^{265}Hs reaction, the cross-section maximum of (51^{+46}_{-29}) pb was obtained at the bombarding energy of 220.5 MeV at the middle of the target. For even-even nucleus ^{264}Hs, we have also observed accurate decay data with good statistics.

3B13 Production and Decay Properties of ^{266}Bh and its daughter nuclei

MORIMOTO, K.[1], MORITA, K.[1], KAJI, D.[1], HABA, H.[1], OZEKI, K.[1], KUDOU, Y.[1], SATO, N.[1,2], SUMITA, T.[1,3], YONEDA, A.[1], ICHIKAWA, T.[1,4], FUJIMORI, Y.[5], GOTO, S.[6], IDEGUCHI, E.[7], KASAMATSU, Y.[1], KATORI, K.[1], KOMORI, Y.[8], KOURA, H.[2], KUDO, H.[6], OOE, K.[8], OZAWA, A.[9], TOKANAI, F.[5], TSUKADA, K.[2], YAMAGUCHI, T.[10], YOSHIDA, A.[1]

([1]Nishina Center for Accelerator Based Science, RIKEN, [2]Japan Atomic Energy Agency, [3]Tokyo University of Science, [4]Yukawa Institute for Theoretical Physics, [5]Yamagata Univ., [6]Niigata Univ., [7]Center for Nuclear Study, Univ. of Tokyo, [8]Department of Chemistry, Osaka University, [9]Univ. of Tsukuba, [10]Saitama Univ.)

A nuclide, ^{266}Bh, is the great-grand-daughter of ^{278}113 that is produced in the ^{209}Bi + ^{70}Zn reaction. The identification was based on a genetic link to the known daughter nucleus ^{262}Db by alpha-decays. The identification of the heaviest nuclides is very difficult because of their extremely small production cross sections. Only five atoms have been assigned to ^{266}Bh by direct production. The main purpose of this work is to provide further confirmation of the production and identification of the isotope ^{278}113.

We have observed 32 correlations in total. 14 correlation events were clearly assigned to the decay chain of ^{266}Bh, and 10 correlations events were tentatively assigned to the decay chain of ^{266}Bh. And the rest correlations were assigned to decay chain of ^{267}Bh or not assigned.

As a present result, a state in ^{266}Bh, which decays by an alpha emission with the energies ranging from 9.05 to 9.23 MeV, feeds a state in ^{262}Db, which decays by alpha emission and by SF with a previously known half-life. The result provided a further confirmation of the production and identification of the isotope of the 113th element, ^{278}113, studied by a research group at RIKEN.