

English Program

2006 Annual Meeting of the Japan Society of Nuclear and Radiochemical Sciences (JNRS) / the 50th Symposium on Radiochemistry

SCIENTIFIC PROGRAMME

Tuesday October 24th

Registration 9:00 -

Room A

9:30 - 10:10

Opening session

Z. Yoshida

K. Kondo

Plenary lecture

1S01 H. Nakahara

Opening address

Welcome address

At the occasion of celebrating the 50th anniversary of the radiochemistry symposium in Japan

10:10 - 12:20

Public session

1S02 M. Imamura

1S03 T. Nakanishi

1S04 M. Ebihara

1S05 Y. Nagame

Science that connects the past and the future - Fifty years of ¹⁴C dating

Water and inorganic elements in plants

Future perspective of neutron-using elemental analysis and its application to cosmochemistry

Elements at the uppermost end of the periodic table

13:20 - 4:20

Memorial session for Professor Tomota Nishi

Oral presentations

14:30 - 18:30

1A01 Y. Takahashi

1A02 H. Ozaki *et al.*

1A03 S. Nagao *et al.*

1A04 N. Tanaka *et al.*

1A05 Y. Oura *et al.*

1A06 K. Shozugawa *et al.*

1A07 M. Segawa *et al.*

1A08 Y. Okada *et al.*

1A09 H. Tagi *et al.*

1A10 K. Tagami *et al.*

1A11 K. Nagai *et al.*

1A12 Y. Kurihara *et al.*

1A13 T. Yamagata *et al.*

1A14 M. Yamada *et al.*

Local structure of rhenium and radiogenic osmium in molybdenite

High-precision wiggle-matching ¹⁴C-dating using Japanese local calibration curve

Study on sources of organic materials in river waters from Bekanbeushi mira by radiocarbon

Determination of siderophile elements in rock samples by using pre-concentration neutron activation analysis

Neutron activation analysis using k₀ standardization method (II)

Analysis of Mn in the pelagic sea sediments by neutron activation analysis and XAFS

2D elemental analysis approach in focused neutron beam induced prompt gamma-ray analysis at JAEA

Determination of trace elements in Katana (Japanese swords) by neutron activation analysis with multidimensional γ-ray spectrometry

Charged particle activation analysis of nitrogen containing 10⁻¹⁴ cm⁻³ order in high purity silicon

Elution behavior of U from a TRU resin cartridge

Activity ratio of Ra isotopes leached from monazite

²³⁰Th/²³⁴U activity ratio in the products from Izu-Bonin island-arc volcanoes

Seasonal variation for Be isotopes in the atmosphere at Tokyo and Hachijo-Island.

¹³⁷Cs inventories in the water column collected from the Sulu and South China Seas

18:45 - 20:15

Division meeting - Nuclear analytical techniques

Room B

12:20 - 13:20

Meeting of young scientists

14:30 - 18:30

Oral presentations

1B01 K. Komura *et al.*

New phenomena - Suppression of gamma rays from radon daughters by phosphomolybdate complex -

1B02 I. Nishinaka <i>et al.</i>	Correlation between fission fragments and α -particles in the $^{18}\text{O} + ^{244}\text{Pu}$ reaction
1B03 H. Haba <i>et al.</i>	Development of a novel heavy element chemistry apparatus using the RIKEN gas-filled recoil separator GARIS as a pre-separator
1B04 H. Kikunaga <i>et al.</i>	Measurement of ^{255}No via the $^{238}\text{U}(^{22}\text{Ne}, 5n)$ reaction for the heavy element chemistry using GARIS as a pre-separator
1B05 Y. Ishii <i>et al.</i>	Cation exchange behavior of Rf in HNO_3/HF mixed solution system (II)
1B06 K. Ooe <i>et al.</i>	Solvent extraction behavior and electrolytic redox reaction of tungsten under extremely low concentration
1B07 T. Ikezawa <i>et al.</i>	Application of mass spectrometry for the identification of chemical species in gas phase chemistry
1B08 H. Ishikawa <i>et al.</i>	Studies on hot atom chemical behavior of energetic ions in solids (IX) - Effects on helium ion implantation for chemical behavior of deuterium implanted into silicon carbide -
1B09 Y. Kikuchi <i>et al.</i>	Studies on hot atom chemical behavior of energetic ions in solids (X) - Ion fluence dependence on chemical behavior of energetic deuterium implanted into oxygen-contained boron film -
1B10 T. Suda <i>et al.</i>	Studies on hot atom chemical behavior of energetic ions in solids (XI) - The dependence of implantation temperature on the chemical behavior of energetic deuterium implanted into highly oriented pyrolytic graphite -
1B11 T. Toraishi <i>et al.</i>	A remote valency control technique for actinides by an external ultrasound irradiation
1B12 A. Kirishima <i>et al.</i>	Study of actinide complexation by calorimetry
1B13 S. Miyashita <i>et al.</i>	Separation of trivalent actinide from lanthanide by a solvent extraction technique using imidazoledithiocarboxylic acid
1B14 T. Yaita <i>et al.</i>	Resonant x-ray emission study on the electronic structure of Cm compounds

18:45 - 20:15

Division meeting – Nuclear chemistry

Wednesday October 25th

Room A

9:10 - 11:55

Oral presentations

2A01 N. Matsuzawa <i>et al.</i>	Variation in the atmospheric concentrations of radioactive lead isotopes with varying meteorological conditions at Kawasaki
2A02 C. Lee <i>et al.</i>	Development of the uranium-enrichment screening method for particles by chemical etching of fission track detector
2A03 H. Kakiuchi <i>et al.</i>	Development of organically bound tritium determination method by noble gas mass spectrometry
2A04 Y. Hamajima <i>et al.</i>	Energy distributions of environmental neutron in liquids and solids
2A05 H. Matsumura <i>et al.</i>	The KENS shielding experiment for high energy neutrons
2A06 K. Bessho <i>et al.</i>	Chemical properties of Be-7 and C-11 formed in the cooling-water of magnets at high-energy proton accelerator facilities
2A07 S. Higaki <i>et al.</i>	Effect of oxygen on the CH_3T and CH_4 emissions from stainless steel surface
2A08 T. Ikai <i>et al.</i>	Synthesis of molecular hexatechnetium clusters by means of dimensional reduction of their polymeric complexes
2A09 M. Kubota <i>et al.</i>	Relationship between inhibiting effect on lipid peroxidation induced by tritiated water and change the shape of lipid membrane due to tea catechins
2A10 K. Washiyama <i>et al.</i>	Biodistribution of ^{224}Ra and ^{223}Ra and retention of its progeny (2)

12:00 - 13:30

Division meeting - Environmental radioactivity

Room B

9:10 - 11:55

Oral presentations

2B01 K. Ninomiya <i>et al.</i>	Electron arrangement of pionic atoms by measuring electronic KX-ray energies
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- 2B02 W. Sato *et al.* Nuclear relaxation and electronic state of ^{140}Ce implanted in highly oriented pyrolytic graphite
- 2B03 S. Tsutsui *et al.* Mössbauer spectroscopy and nuclear resonant forward scattering of $\text{RRu}_4\text{P}_{12}$ (R=rare earth metal)
- 2B04 Y. Nakajima *et al.* Mössbauer spectroscopic studies of alkylammonium iron complexes
- 2B05 J. Sakuma *et al.* Mössbauer studies and magnetic susceptibility of SnO_2 doped ^{57}Fe
- 2B06 S. Hayami *et al.* Mechanism of Iron(III) LIESST Complexes
- 2B07 S. Nakashima *et al.* Spin-crossover phenomena of the assembled complexes and their Mössbauer spectra
- 2B08 T. Nishida *et al.* Mössbauer spectra of new cathode material for lithium ion battery
- 2B09 K. Oshida *et al.* Photocontrol of the electronic states of iron coordinated to photoresponsive polyaniline
- 2B10 Y. Sakai *et al.* Effective application of Mössbauer spectroscopy to chemical education - Support to general chemistry laboratory of redox reactions of iron -

12:00 - 13:30

Division meeting - Nuclear processes as chemical probes

Room A

13:40 - 15:10

Invited lectures

2S01 A. Chatt Radiochemistry research and training in Canada - Past, present and future -

2S02 Y. F. Liu Current development of nuclear power plants, nuclear waste geological disposal and nuclear education in China

2S03 Y. Maeda Present situation and future prospects of the Japan Society of Nuclear and Radiochemical Sciences

15:30 - 16:20

Plenary lecture

2S04 D. C. Hoffman Past, present and future of nuclear and radiochemistry in the U. S.

16:20 - 17:10

Plenary lecture

2S05 N. Saito Dawn of radiochemical research in Japan - From Hokutolite to U-237 -

17:10 - 17:30

Commendation ceremony

18:00 - 20:00

Celebration of the 50th anniversary of the radiochemistry symposium (Mito Keisei Hotel)

Thursday October 26th

Room E

9:30 - 12:10

ASR2006 (<http://asrc.tokai-sc.jaea.go.jp/ASR2006/index.html>)

Room C

13:15 - 14:30

General meeting of JNRS

The JNRS award ceremony

14:40 - 15:20

Awardees lecture of the JNRS award

3S01 T. Ohtsuki Enhanced electron-capture decay rate of ^7Be encapsulated in C_{60} cages and their electron structure

15:20 - 16:20

Awardees lectures of the JNRS award for young scientists

3S02 A. Kitatsuji Electrochemical study of the ion transfer reaction at liquid / liquid interface and application to actinide separation

3S03 T. Suzuki Separation of lanthanides and actinides by using tertiary pyridine resin

Room E

16:30 - 18:30

ASR2006

Room D

16:30 - 18:30

Poster presentations

- 3P01 M. Fukushima *et al.* Multi-element analysis of cultivated oyster tissues from Miyagi
- 3P02 S. Kasahara *et al.* A study on the distribution of cobalt in Clethrionomys leaves by solvent extraction method with supersonic
- 3P03 M. Kawashima *et al.* Determination of concentrations of trace elements in pancreatic cell of zinc deficient mice
- 3P04 J. Kamishima *et al.* Variation in concentrations of trace elements and metalloproteins in hepatocyte of zinc deficient mice
- 3P05 D. Moromachi *et al.* Environmental evaluation of tidal sediments by neutron activation analysis and Moessbauer spectroscopy
- 3P06 M. Kawano *et al.* Determination of Extractable Organohalogens (EOX) by neutron activation analysis and individual organohalogen compounds by GC-MS in soil and sediment samples
- 3P07 Y. Hatsukawa *et al.* Study on Presolar grains using multiparameter coincidence method
- 3P08 Y. Takata *et al.* An attempt to determine trace elemental abundances in seawater by INAA using a multipul gamma-ray detector.
- 3P09 K. Washiyama *et al.* Evaluation of activable tracer for nuclear medicine
- 3P10 K. Komoto *et al.* Stability of ^{186}Re - and ^{188}Re -DMSAs and removal of impurity in the product
- 3P11 S. Ishioka *et al.* Simultaneous production of high-pure ^{76}Br , ^{77}Br and ^{79}Kr using an isotope separator
- 3P12 M. Sakama *et al.* Determination of the neutron flux at the self-shielded PET cyclotron of the University of Tokushima using the activation foil method
- 3P13 Y. Hakuma *et al.* Oxidative stress by Se-deficiency and dynamics of biotrace elements
- 3P14 M. Kubota *et al.* Study on inhibition mechanism of tea catechins on beta-ray-induced lipid peroxidation by examination shapes of liposome
- 3P15 T. Sato Activity ratio of Pm-146/Pm-147 in Pm-147 radiation source
- 3P16 Y. Endo *et al.* Study on Hf metallofullerenes using ^{181}Hf radioactive-tracer (2)
- 3P17 S. Watanabe *et al.* Synthesis of endohedral ^{133}Xe -fullerenol by using higher fullerene
- 3P18 A. Fujiwara *et al.* Study on the analytical method of $^{242\text{m}}\text{Am}$ in solidified products
- 3P19 R. Doi *et al.* Modeling on Cs sorption behavior onto sedimentary rock
- 3P20 M. Kamoshida *et al.* Separation of uranium using microcapsule of tri-n-octylphosphine oxide
- 3P21 T. Kuribayashi *et al.* The development of expeditious and high efficient separation method of tervalent actinoids using α -HIBA as a ligand
- 3P22 T. Mori *et al.* Separation of trivalent actinides from lanthanides using a capillary electrophoretic technique
- 3P23 R. Idobata *et al.* Study on f-block element ions by solvent extraction using cyclic polythiaether
- 3P24 T. Suzuki *et al.* Adsorption behavior of f-elements on tertiary pyridine and quaternary pyridinium resins
- 3P25 H. Kubota *et al.* Determination of transfer energy of actinide ions between aqueous and organic solution phases
- 3P26 Y. Sugo *et al.* Solvent extraction of actinide ions by *N,N*-dioctyldiglycolamic acid
- 3P27 M. Watanabe *et al.* Separation of Am(III) by nitrogen-donor ligand
- 3P28 T. Kawasaki *et al.* Synthesis and crystal structure of various Uranyl(VI) complexes coordinated by acac
- 3P29 K. Hayashi *et al.* The interaction between trivalent f-block element ions and iodide ion
- 3P30 J. Komagine *et al.* Preparations and crystal structures of 8 coordinate uranyl (VI) complexes having macrocyclic ligands derived from pyrroledicarboxialdehydes and diamines.
- 3P31 T. Kakuyama *et al.* Positive muons in β -tricalcium phosphate
- 3P32 T. Nakatsuka *et al.* Basic study for the gas-phase chemical reaction of muonic atoms
- 3P33 H. Sakai *et al.* Mössbauer spectra of tris(2-pyridylmethyl)amine-iron complexes
- 3P34 S. Iijima *et al.* Spin crossover behaviors of Fe(II) complexes of polydentate ligands containing imidazole group

3P35 K. Hiki <i>et al.</i>	Photoinduced spin transition phenomena for Fe(III) spin-crossover complex
3P36 Y. Einaga <i>et al.</i>	Photo-control of magnetization in magnetic nano-particles
3P37 K. Nagata <i>et al.</i>	Characterization of iron-nanoparticles in carbon nanotubes by Mössbauer spectroscopy
3P38 Y. Yamada <i>et al.</i>	Effects of droplets on spin-orientation of iron films produced by laser-deposition
3P39 Y. Tsuruoka <i>et al.</i>	In-beam Mössbauer spectroscopic study on the products of neutron capture reaction in iron disulfide
3P40 K. Nakagami <i>et al.</i>	Mössbauer resonance absorption on FePSe ₃ and FePTe ₃
3P41 Y. Suzuki <i>et al.</i>	Mössbauer study on IZO doped with diluted Fe
3P42 K. Garcia <i>et al.</i>	On the formation of akaganeite in the presence of different cations
3P43 M. Takahashi <i>et al.</i>	Iodine-127 Mössbauer spectra of (dihaloiodo)benzenes
3P44 T. Ayabe <i>et al.</i>	¹⁵⁵ Gd Mössbauer spectroscopic investigation of 3d-4f
3P45 M. Nakada <i>et al.</i>	Synthesis method and Mössbauer spectroscopic study of cyanopyridine coordinated neptunyl complex
3P46 N. Masaki <i>et al.</i>	Mössbauer spectra of U-Eu mixed oxide

Friday October 27th

Room E
9:20 - 18:00
ASR2006

Room D
10:00 - 12:00
Poster presentations
4P01

- withdrawal -

4P02 M. Sakamaki <i>et al.</i>	Development of gridded ionization chamber to measure atomic number of fission fragments
4P03 M. Arai <i>et al.</i>	Study on synthesis of heavy elements using the Ba + ¹⁶ O, La + ¹⁶ O reaction systems
4P04 K. Nakashima <i>et al.</i>	Measurement of ultraviolet, visible and infrared light originating from de-excitation of ^{229m} Th
4P05 H. Koura <i>et al.</i>	Systematics properties of spontaneous fission for superheavy nuclei and a limit of existence of nuclei
4P06 A. Osa <i>et al.</i>	Development of ion source for JAEA-ISOL –I
4P07 K. Sato <i>et al.</i>	Development of ion source for JAEA-ISOL –II
4P08 T. Takabe <i>et al.</i>	Heavy element production at RIKEN for chemistry of seaborgium
4P09 Y. Tashiro <i>et al.</i>	Startup of on-line chemistry at Research Center for Nuclear Physics, Osaka University
4P10 Y. Tashiro <i>et al.</i>	Rapid solvent extraction experiment of No ²⁺ with multi-track microchips
4P11 T. Ishikawa <i>et al.</i>	Basic study on gas phase chemistry using carbon cluster transport system
4P12 A. Toyoshima <i>et al.</i>	Development of electrochemistry apparatus for atom-at-a-time chemistry
4P13 A. Toyoshima <i>et al.</i>	TOPO reversed-phase extraction behavior of rutherfordium in HCl solutions
4P14 Y. Kasamatsu <i>et al.</i>	Adsorption of Nb, Ta and Pa on anion exchangers in diluted HF medium
4P15 K. Akiyama <i>et al.</i>	EXAFS Study for complexation of Zr and Hf with TBP in HCl solution
4P16 T. Yoshida <i>et al.</i>	Elucidation of accumulation mechanism ¹⁰ Be to bottom of the ocean with sediment
4P17 M. Yamamoto <i>et al.</i>	Variation of uranium and thorium isotopes in Lake sediments-Lake Hovsgol in Mongolia
4P18 T. Nakano-Ohta <i>et al.</i>	Determination of ²²⁶ Ra and ²²⁸ Ra in surface seawater around Sagami Bay and its vicinity
4P19 T. Aono <i>et al.</i>	Vertical profiles of thorium isotopes with the multiple-unit large-volume <i>in situ</i> filtration system in the North Pacific

- 4P20 Y. Sato *et al.* A study of high yield separation of trace plutonium isotopes from manganese crust
- 4P21 J. Zheng *et al.* Determination of Pu isotopes in settling particles at femtogram levels using SF-ICP-MS with a high-efficiency sample introduction system
- 4P22 S. Kihara *et al.* $^{239,240}\text{Pu}$ inventories in marine sediments in the Pacific waters near Japan
- 4P23 T. Izumi *et al.* Depth distribution of ^{241}Am in water columns of the Eastern Indian Ocean and its adjacent sea
- 4P24 T. Saito *et al.* Radioactivity of uranium-series and thorium-series nuclide in Tamagawa hot-spring water Akita prefecture, Japan.
- 4P25 Y. Kanai *et al.* Variation of uranium-series nuclides in borehole groundwaters from Kanamaru, Yamagata Prefecture
- 4P26 A. Hirose *et al.* Environmental tritium dynamics model in a small drainage basin
- 4P27 M. Takahashi *et al.* $^{230}\text{Th}/^{234}\text{U}$ activity ratio in rhyolite from Izu-Mariana arc
- 4P28 Y. Yoshizawa *et al.* Estimation of effective dose from Rn emanating from the "minus ion" effect wallpaper
- 4P29 T. Abe *et al.* High resolution simultaneous measurements of airborne radionuclides
- 4P30 Y. Yamaguchi *et al.* High resolution simultaneous measurements of airborne radionuclides in the pan-Japan sea area
- 4P31 H. Namioka *et al.* Characterization of fallout deposition materials during 1993-2005 at Tokai-mura by radioanalytical methods
- 4P32 I. Morinaga *et al.* Seasonal variations of the source of aerosol particles by using backtrajectories and principal component analysis
- 4P33 T. Esaka *et al.* Development of detection method for individual environmental particles containing alpha radioactive nucleus
- 4P34 M. Inoue *et al.* Measurement of radionuclides contaminated in chemical reagents
- 4P35 H. Sakaue *et al.* On-line monitoring of radioactive dust using high-speed pulse interval analysis for multi-input channels
- 4P36 K. Yasuda *et al.* Instrumental development for measurement of elements distribution on the surface of smear samples.
- 4P37 Y. Saito-Kokubu *et al.* Plutonium in the Nishiyama area of Nagasaki city
- 4P38 Y. Inoue *et al.* Obstruction of fluorescence X-ray emitted from Sm to the determination of ultra-low-level ^{152}Eu separated from a sample exposed to atomic bomb
- 4P39 T. Imanaka *et al.* Radiation dose due to neutron-induced residual radioactivities by the atomic bombs in Hiroshima and Nagasaki
- 4P40 Y. Ohtsuka *et al.* Concentrations of ^{99}Tc and ^{137}Cs in edible kelps and sea urchin ovaries from the northern part of Japan
- 4P41 H. Kawamura *et al.* Method for measurement of ^{129}I in plant samples by AMS
- 4P42 Y. Takada *et al.* Depth profile of ^{129}I concentrations in soil determined by AMS
- 4P43 N. Yamaguchi *et al.* Relationships between the long-term mobility of ^{90}Sr in ploughed soil of upland field and soil properties.
- 4P44 Y. Miyamoto *et al.* Trace analysis of uranium, thorium, lead and lanthanoid in environmental samples - Examination of condition of anion-exchange separation (II) -
- 4P45 N. Osada *et al.* Formation of radioactive aerosol in an electron linear accelerator irradiation room

Room D

12:10 - 12:30

Closing session

14:00 - 16:00

Technical tour of J-PARC (in Japanese)

English Abstracts

1S01 : At the occasion of celebrating the 50th anniversary of the radiochemistry symposium in Japan

Nakahara, H. (Tokyo Metropolitan Univ.)

We are pleased to announce that the radiochemistry symposium to be held at Mito and Tokai this year is the 50th symposium since Prof. Nobufusa Saito organized the first one in 1957. It has been held at various universities or research institutes by distinguished leaders of the region, and the symposium has been organized under the joint auspices of the Chemical Society of Japan, the Atomic Energy Society of Japan, and the Japan Society for Analytical Chemistry, and more recently by The Pharmaceutical Society of Japan. Their continued support is, here, gratefully acknowledged.

For this memorial occasion, we, organizers, have planned several special events listed below to visit the past of our predecessors and to hand over their precious experience to our brilliant and hopeful successors of the next generation. (1) We prepare sessions in the symposium which are open to the public and with special invited talks by the world top-notch radio- and nuclear chemists who will disclose to us the present status and the future of radiochemistry. (2) Two kinds of books are now under preparation: one is "The course of history of radiochemistry in Japan" and the other is "A dictionary of radiochemistry terminologies". (3) A DVD will be made which contains all the proceedings of 50 radiochemistry symposia. (4) Text books for radiochemistry are being planned by the board of directors of the Japan Society of Nuclear and Radiochemical Sciences that covers from the fundamental to the applied with the hope that they will become invaluable not only for educating young students at universities but also for making the scientists around us to understand what radiochemistry is and how indispensable it is not only for the peaceful use of atomic energy but also for all the sciences related to our daily life, such as medicine, agriculture, pharmacy and archeology.

1S02 : Science that connects the past and the future - Fifty years of ^{14}C dating -

Imamura, M. (National Museum of Japanese History, National Institutes for Humanities)

Major contribution of radiochemistry in archeological sciences is the dating which provides the most essential information in studies of prehistory. In particular the method of ^{14}C dating revolutionized the theory of civilization in the world. In this talk I will review 50 years of ^{14}C dating, techniques and applications, and then the possibilities in contemporary world. The technical developments are featured by ultra-sensitive method of accelerator mass spectrometry in 1977 and its developments, and an introduction of international calibration datasets for ^{14}C dates started in 1986. Both contributed greatly to improvements in versatility and precision for dating. As for the applications, several interesting topics are reviewed such as the origin of agriculture and its propagation in the western world, the oldest pottery production in Japanese archipelago and its relation to world climate, paddy-rice agriculture and its propagation in Japanese archipelago. Since ^{14}C studies in nature is also well known as key information on carbon dynamics in earth's surface, it may be said that ^{14}C play an extraordinary role in understanding our past, present and the future

1S03 : Water and Inorganic Elements in Plants

Nakanishi, T. (The Univ. of Tokyo)

The author presents features of imaging methods of a living plant using neutron beam, positron emitters as well as conventional radioisotopes for tracer works, such as ^{14}C , ^{45}Ca or ^{32}P . Neutron beam provides water image of a living plant, not only up-ground part of the plant but also roots imbedded in soil. When ^{15}O -water was applied to a plant, it was found that tremendous amount of water was leaking out from xylem vessels and re-entered to the vessels. The advantage of using positron emitters is that the same experiment is able to repeat using the same sample, since half-lives of most of positron emitters are short. However, there is a serious problem of "positron escape" from the sample, which makes it difficult to determine the tracer amount in the sample. To avoid this problem we developed real-time isotope imaging system using conventional beta-emitters, composed of a solid scintillator and a photon counter. This system could provide an image, which sensitivity is more than 10 times higher than that of an imaging plate and was able to analyze the isotope movement from one cell to the other.

1S04 : Future perspective of neutron-using elemental analysis and its application to cosmochemistry

Ebihara, M. (Tokyo Metropolitan Univ.)

Neutron activation analysis is a representative analytical method of elements among not only activation analyses but also analytical methods in general. High sensitivity for many elements can be obtained even in non-destructive analysis. For neutron activation analysis, research reactors are the most commonly used for neutron sources. Although the number of research reactors usable for neutron activation analysis has been decreased for last few decades, its applicability seems to have been anti-proportionally expanding. Prompt gamma ray analysis must be an example of showing such increasing applicability; users number is rapidly expanding and its contribution to science is growing. In a few years, a new accelerator facility for producing neutrons by a spallation reaction will become available. We understand that such neutrons cannot be used in the same manner as those provided by research reactors but we believe that a new page for neutron activation will be opened. Advantages of neutron activation analysis can be exhibited the most effectively in analyses of cosmochemical samples. In the lecture, several examples showing effectiveness of neutron activation analysis in cosmochemistry will be also presented.

1S05 : Elements at the upper most end of the periodic table

Nagame, Y. (JAEA)

The elements with atomic numbers $Z \geq 104$ are called transactinide elements. The currently known transactinide elements, elements 104 through 112, are placed in the periodic table under their lighter homologues in the 5d electron series, Hf to Hg. Elements from 113 to 118 except for 117 that have been recently synthesized would be in the successive 7p electron series. Studies of chemical properties of the transactinide elements offer unique opportunities to obtain information about trends in the periodic table of the

elements at the limits of the nuclear stability. In this report, the recent progress of chemical studies of the transactinide elements as well as the synthesis of these elements in Japan is briefly summarized.

2S01 : Radiochemistry research and training in Canada - past, present and future -

Chatt, A. (Dep. of Chem., Dalhousie Univ., Canada)

Canada had a fairly good start in radiochemistry. We had high-flux research reactors at Chalk River Nuclear Laboratories and McMaster University, a cyclotron at McGill University, and electron linear accelerators (Linac) at the University of Saskatchewan (UoS) and University of Toronto (UoT). Both the number of trainees and research projects in radiochemistry exploded by mid-1970's with the installation of SLOWPOKE-2 research reactors at UoT, Ecole Polytechnique, Dalhousie University (DU), University of Alberta, Saskatchewan Research Council, and Royal Military College of Canada. These facilities are mainly involved in NAA and radiochemical separations. Then TRIUMF was commissioned at Simon Fraser University mainly for isotope production. In the meantime the UoT Linac, and AECL-CP and UoT SLOWPOKE-2 reactors were shut down. But a third generation synchrotron facility called the Canadian Light Source (CLS) at UoS came into operation. The School of Energy Systems and Nuclear Science at the University of Ontario Institute of Technology (UOIT) is interested in starting up a new radiochemistry program. The role of radiochemistry in nanoparticle research and in radiological and nuclear counter terrorism response has become very important. More funding is becoming available to establish more radiochemical laboratories, develop new methodologies, do more research, and train more personnel.

2S02 : Current development of nuclear power plants, nuclear waste geological disposal and nuclear education in China

Liu, Y. F. (College of Chem., Peking Univ., China)

See original abstract (2S02)

2S03 : Present situation and future prospects of the Japan Society of Nuclear and Radiochemical Sciences

Maeda, Y. (Kyushu Univ.)

I would like to congratulate the Symposium on Radiochemistry on the occasion of its 50th Anniversary. The Radiochemistry Symposium is an annual meeting on radiochemistry that was founded in 1957 and has contributed to the progress of this field since its inception. The Japan Society of Nuclear and Radiochemical Sciences (JNRS) was founded on October 12, 1999 to revitalize studies on radiochemistry and related fields by the members of the Symposium on Radiochemistry. The historical scope of the society will be reviewed. Among other activities the challenges of global environmental protection must be met to preserve our world for future generations. We must work together to continuously communicate with the relevant societies of other Asian countries to effectively address such

problems.

2S04 : Past, present, and future of nuclear and radiochemistry in the U.S.

Hoffman, D. C. (Dep. of Chem., Univ. of California, Berkeley and LBNL, USA)

See original abstract (2S04)

2S05 : Dawn of radiochemical research in Japan - From Hokutolite to U-237 -

Saito, N. (Prof. Emer., Univ. of Tokyo)

This talk will present important developments of Japanese radiochemical studies in both natural and artificial radioelements with emphasis on two topics. First, the talk will describe radiochemistry of Hokutolite, a radioactive sinter deposit, in Peitou Hot Springs in Taiwan as well as Tamagawa Hot Springs in Japan after the discovery of Hokutolite in 1907. Secondly, the talk will mention Nishina and Kimura's excellent work on the discovery of a new uranium isotope, U-237, and several new fission products produced by fast neutron bombardments of uranium with RIKEN cyclotron during the period 1938-1942.

3S01 : Enhanced electron-capture decay rate of ^7Be encapsulated in C_{60} cages and their electron structure

Ohtsuki, T. (Lab. of Nucl. Sci., Tohoku Univ.)

Decay rate of ^7Be electron capture was measured in C_{60} and Be metal with a reference method. The half-life of ^7Be endohedral C_{60} ($^7\text{Be}@\text{C}_{60}$) and ^7Be in Be metal (Be metal(^7Be)) is found to be 52.65 ± 0.04 and 53.25 ± 0.04 days, respectively. This amounts to a 1.1% difference in EC-decay half-life between $^7\text{Be}@\text{C}_{60}$ and Be metal(^7Be). We also present calculations of the electron density at the ^7Be nucleus at the site inside C_{60} and in Be metal crystal. These values are in fair agreement with the experimental values. Our result is a reflection of the different electron wave-functions for $^7\text{Be}@\text{C}_{60}$ inside C_{60} compared to the situation when ^7Be is in a Be metal.

3S02 : Electrochemical study of the ion transfer reaction at liquid / liquid interface and application to actinide separation

Kitatsuji, Y. (JAEA)

Transfer of actinide ions at the interface between two immiscible electrolyte solutions has been investigated by electrochemical method. In order to elucidate the transfer of actinide ions between two phases, a novel electrochemical method based on controlled-potential electrolysis has been developed. It was found that the method was applicable to the study of the transfer of such hydrophilic ions as transition metal ions which gave no appreciable current within the potential window in voltammetry or polarography at the interface. Controlled-potential electrolysis developed was applied to the transfer reactions of actinide ions such as UO_2^{2+} and Am^{3+} from aqueous solution to organic solution in

the absence or presence of an ionophore facilitating the transfer. Gibbs energy for the transfer of actinide ion and a stability constant of the complex between an actinide ion and the ionophore in organic solution were determined. The controlled-potential electrolysis at the interface between two phases has feasibility as a method for electrolytic separation of actinide ions.

3S03 : Separation of lanthanides and actinides by using tertiary pyridine resin

Suzuki, T. (Tokyo Tech.)

The group separation of lanthanides and actinides and the mutual separation of elements in each group were studied by using tertiary pyridine resin with the hydrochloric acid or the nitric acid / alcohol mixed solution. The tertiary pyridine resin has two functions; one is a function of weakly basic anion exchanger, and another is a function of soft donor ligand. The completely group separation of lanthanides and actinides was confirmed to be achieved in the hydrochloric acid system. It was confirmed that the mutual separation of lanthanides and actinides is possible by using the nitric acid / alcohol mixed solution. It was found that the adsorption of actinides on tertiary pyridine resin and the separation phenomenon of actinides from lanthanides in the hydrochloric acid system are due to the coordinate linkage between cation and nitrogen donor ligand of pyridine and obey the HSAB (hard and soft acids and bases) principle. On the other hand, in the nitric acid system, it was found that the separation is caused by the electrostatic interaction based on the anion exchange.

1A01 : Speciation of rhenium and radiogenic osmium in molybdenite by sensitive XAFS

Takahashi, Y. (Hiroshima Univ.)

For the speciation of trace elements in rocks and minerals, fluorescence XAFS using energy-dispersive Ge detector has been often used. By this method, however, high quality fluorescence XAFS cannot be obtained under intense scattering and/or fluorescence from other predominant elements in the samples. To deal with this problem, we developed a system for fluorescence XAFS using a bent crystal analyzer to selectively extract fluorescence x-rays of a target element. In this paper, speciation of Os in molybdenite has been studied as an example of the application. Radiometric dating using various radioactive decay systems has been widely applied to various terrestrial and extraterrestrial materials. Although the information is closely related to chemical stabilities of parent and daughter nuclides and the reliability of the radiometric dating, there have been few investigations on the direct identification of the chemical state of the daughter nuclides. In this study, we chose Re-Os system in molybdenite for the possible application of this idea, since initial abundance of Os in molybdenite is often negligible compared with radiogenic Os in molybdenite rich in Re. From XAFS spectra, we have investigated local structure of Re and Os in molybdenite. Details of the analyses will be given in the presentation.

1A02 : High-precision wiggle-matching ^{14}C -dating using Japanese local calibration curve

Ozaki, H.,¹ Imamura, M.,¹ Matsuzaki,² H., Nakamura, T.,³

Niu, E.,⁴ Itoh, S.,⁴ Mitsutani, T.⁵ (¹National Museum of Japanese History, ²Dep. of Nucl. Eng. and Management, Univ. of Tokyo, ³Center for Chronological Res., Nagoya Univ., ⁴Paleo-Labo Co., ⁵National Res. Inst. for Cultural Properties, Nara)

International radiocarbon calibration curve so called IntCal has been constructed based on ^{14}C concentrations of samples dated by other dating methods. Regional offsets of ^{14}C concentration from IntCal have been pointed out frequently based on the some differences found in samples from different regions. Although ^{14}C wiggle-matching technique is effective to obtain precise dates, regional offsets may make systematic errors in results of ^{14}C wiggle-matching. In this study, we investigate a local calibration curve for Japanese samples using Japanese tree-ring samples and apply ^{14}C wiggle-matching using Japanese local calibration curve.

1A03 : Study on source of organic materials in river waters from wetland by radiocarbon

Nagao, S.,¹ Aramaki, T.,² Kodama, H.,³ Tani, M.,⁴ Kumegawa, M.,¹ Shibata, Y.² (¹Hokkaido Univ., ²NIES, ³Kyoto Pref. Univ., ⁴Obihiro Univ. Agr. Veterinary Medicine)

Radioactive and stable isotopes of carbon (^{14}C and ^{13}C) can serve as powerful tools for identifying sources and estimating turnover times of organic matter in aquatic systems. This study is presented for application of ^{14}C and ^{13}C to the transport of particulate and dissolved organic materials in river waters from the Betsu River in Japan. Samples for the surface river water were collected at a station on summer (July in 2004 and August in 2005) and early winter season (November in 2003 and December in 2005). The ^{14}C measurements were performed by accelerator mass spectrometry (AMS) at the National Institute for Environmental Studies. The $\Delta^{14}\text{C}$ value is +9.2‰ for organic matter in suspended particles in July. The $\Delta^{14}\text{C}$ values of fulvic acids are -30‰ and -12.2‰ for the winter samples, and +9.7‰ for the summer sample. The $\delta^{13}\text{C}$ values range from -30.8 to -27.4‰. These results suggest that the transport of dissolved and particulate organic matter very with season

1A04 : Determination of siderophile elements in rock samples by using pre-concentration neutron activation analysis

Tanaka, N., Oura, Y., Ebihara, M. (Grad. School of Sci. and Eng., Tokyo Metropolitan Univ.)

Nickel sulfide (NiS) fire assay method is often used as pre-concentration technique for platinum group elements (PGEs) and gold in rock samples. Besides these elements, NiS beads are expected to contain other siderophile and chalcophile elements. We performed neutron activation analysis for NiS beads directly and tried to determine any elements concentrated in beads including PGEs and gold. The critical factor affecting γ -ray spectrometry is induced radioactivity of ^{58}Co derived from the reaction of ^{58}Ni (n, p) ^{58}Co . To reduce this interference, fire assay process was performed with smaller amount of Ni powder. Considering radioactivity of ^{58}Co , we selected two irradiation sites which have relatively high Cd ratios. The results of measurement and radioactive tracer experiment supported not only PGEs but also some of chalcophile elements like arsenic, selenium

and antimony were concentrated in beads. Analytical results for Ir and Au were in agreement with literature values even for the small scaled fire assay process. Radioactivity of ^{58}Co was significantly decreased in the case of irradiation at JRR-3M PN3 (Cd ratio= 280).

1A05 : Neutron activation analysis using k_0 standardization method (II)

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

The k_0 -IAEA, a software developed by IAEA for neutron activation analysis using k_0 standardization method, was opened in 2005, and have been developing. The present k_0 -IAEA software includes a procedure for a calculation of peak areas in gamma-ray spectra. Using only k_0 -IAEA, we can obtain concentration values now. We present an experience and an accuracy of concentration values by k_0 -IAEA based on our use. Peak area values calculated by k_0 -IAEA were compared with those by HypermetPC and WPKarea software. For values larger than 10000 counts, peak areas calculated by three each software were consistent, but values were different by several 10s% from each other for peak area smaller than 10000 counts. Concentration values obtained by k_0 -IAEA for SRMs, SMELTS Type II and Type III were tended to be 10% - 20% lower than reference values. It is guessed that correction for counting efficiency and sum effect for geometry of samples are not unsatisfactory in the k_0 -IAEA software.

1A06 : Analysis of Mn in the pelagic sea sediments by neutron activation analysis and XAFS

Shozugawa, K.,¹ Sano, Y.,² Matsuo, M.¹ (¹Grad. School of Arts and Sci., The Univ. of Tokyo, ² Ocean Res. Inst., The Univ. of Tokyo)

In order to estimate the early stage of diagenesis of the pelagic sea sediments, we measured the concentrations of Mn and other elements by INAA and chemical states of Mn by XAFS. Pelagic sea sediment, collected from South Pacific Ocean (S39°59.57', W169°59.85', depth: 4627m), had been kept a fine condition that had no disconcertion when it excavated. And the core length is about 30cm. Concentration of Mn was 0.25% entire the core, suggesting that the stage of O_2 reduction terminated. In the RSF spectrum by XAFS measurements, as the depth increases (0.5-28.5cm), the strength of Mn-O tends to decrease continuously. This tendency cannot be observed in the core which had the long scale of sedimentation age. The fact of decrease of Mn-O intensity indicates the possibility of the progress of Mn-reduction occurred in the sediments.

1A07 : 2D Elemental analysis approach in focused neutron beam induced prompt gamma-ray analysis at JAEA

Segawa, M.,¹ Matsue, H.,¹ Sekiya, Y.,² Yamada, S.,¹ Shinohara, T.,¹ Oku T.,¹ Sasao, H.,² Suzuki, J.,¹ Shimizu, H. M.³ (¹JAEA, ²RADA, ³KEK)

Since neutron has the stronger penetration power than those for charged particles, the 2 dimensional prompt gamma-ray analysis method with use of neutron beam is the only way enables the nondestructive, isotopic and position dependent observation on small quantities of samples. Hence,

we aimed at developing 2 dimensional prompt gamma-ray analysis systems in focused neutron beam at JAEA with a high position resolution within a 1mm. A 2-demeinsinal analysis system is set at JRR-3 prompt gamma-ray system. The neutron beam is 0.6mm FWHM focused and collimated with use of a multichannel parabolic guide and a ^6LiF collimator set at 90cm and 1.5 cm upstream with respect to the neutron beam direction from a sample position, respectively. The sample could be stepping drive with 0.1mm. Finally, we succeed to increase the position resolution within 1mm, since we could split two neutron captured gamma-ray peaks from two minute Cd wire-sample, the diameter is 0.25mm ϕ and the length is 5 mm, set at 1mm intervals.

1A08 : Determination of trace elements in Katana (Japanese sword) by neutron activation analysis with multidimensional γ -ray spectrometry

Okada Y.,¹ Hirai S.,¹ Ohya S.,¹ Kimura A.,² Hatsukawa Y.,² Toh Y.,² Koizumi M.,² Oshima M.² (¹Musashi Inst. of Tech., ²JAEA)

In this study, we tried to measure trace elements (As and Sb) in Katana (Japanese swords) by multidimensional γ -ray spectrometry method (GEMINI-II) and conventional counting method for neutron activation analysis (NAA). The determined values by GEMINI-II and conventional counting were in good agreement with. Using the multidimensional γ -ray spectrometry (GEMINI-II) to determine As and Sb was improved by 7 times and 10 times compared with the conventional counting method.

1A09 : Charged particle activation analysis of ppb level nitrogen in high purity silicon

Yagi, H.¹, Masumoto, K.², Minai, Y.³, Nozaki, T.⁴, Saito, Y.⁵, Inoue, N.⁶ (¹SHIEI, ²KEK, ³Musashi Univ. ⁴Purex, ⁵JRIA, ⁶Osaka Pref. Univ.)

It is important to control ultra trace level of nitrogen in silicon for improving the performance of Si crystals. We try to examine and improve the reliability and accuracy of charged particle activation analysis. We adopted the $^{14}\text{N}(p,\alpha)^{11}\text{C}$ reaction for the activation of nitrogen, and the behavior of ^{11}C in its chemical separation from the bombarded silicon was examined in two separation procedures, dry fusion and wet chemistry, in two cyclotron facilities. Nitrogen of 10^{14} atoms/cm³ level in CZ silicon has proved to be determined by both methods, which gave results agreeing fairly well with each other and with the result of SIMS.

1A10 : Elution behavior of uranium from a TRU resin cartridge

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

In order to separate and concentrate uranium from environmental samples for determination of $^{235}\text{U}/^{238}\text{U}$ isotopic ratio by quadrupole ICP-MS, a TRU resin procedure was studied. A water sample (100-200 mL), acidified to 1M HNO_3 , was passed through the resin cartridge at 4-5 mL/min flow rate to extract U in the solution onto the resin, then, the cartridge was washed with 0.5M HCl. Most of the matrix elements in the sample solution were not retained on the resin when the sample solution was passed through, and all Th and some portions of the rare earth elements were eluted from the

resin with the wash solution. For removal of U from the resin cartridge, 0-0.07M tetramethyl ammonium hydroxide (TMAH) was used. No elution of U with deionized water was observed, however, 10 mL of 0.07M TMAH could extract almost all the U from the resin cartridge.

1A11 : Activity ratio of Ra isotopes leached from monazite

Nagai, K., Sato, J. (Meiji Univ.)

The activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ in leachate from monazite was observed to decrease with increasing in pH of the solution, and the activity ratio of $^{224}\text{Ra}/^{228}\text{Ra}$ in leachate increased. ^{226}Ra formed through 3α - and 2β -decays from ^{238}U and ^{224}Ra formed through 2α - and 2β -decays from ^{232}Th tended to be leached more effectively than ^{228}Ra formed through a single α -decay from ^{232}Th . The leaching of Ra isotopes may possibly be dependent on the difference in the history of decay processes which have formed Ra isotopes.

1A12 : $^{230}\text{Th}/^{234}\text{U}$ activity ratio in the products from Izu-Bonin island-arc volcanoes

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

Magma genesis at 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 slab. Uranium-series disequilibria in the volcanic products are a useful tracer to understand various magma processes. Young island-arc volcanic rocks showed the characteristic feature of excess ^{234}U over ^{230}Th . Observation was carried out on the radioactive disequilibrium between ^{234}U and ^{230}Th in the volcanic products from Asama volcano and Izu-Mariana island-arc volcanoes. Thorium and Uranium in the volcanic rock samples were separated by anion-exchange resin and purified by TEVA-Spec. and UTEVA-Spec. resins, respectively. Purified Th and U were electrodeposited onto a stainless steel planchet for α -ray counting. U-234 and ^{230}Th in volcanic rock samples were determined by isotope dilution method coupled with α -ray spectrometry. $^{230}\text{Th}/^{234}\text{U}$ activity ratio in the volcanic products from Asama volcano and Izu-Bonin island-arc volcanoes were in radioactive disequilibrium, enriched in ^{234}U relative to ^{230}Th , which is often observed for volcanic products from young island-arc volcanic products.

1A13 : Seasonal variation for Be isotopes in the atmosphere at Tokyo and Hachijo-Island

Yamagata, T.¹, Saito, T.², Matsuzaki, H.³, Nagai, H.² (¹Grad. School of Integrated Basic Sci., Nihon Univ., ²College of Humanities and Sci., Nihon Univ., ³School of Eng., The Univ. of Tokyo)

The concentration of cosmogenic nuclides ^7Be ($T_{1/2}=53.3\text{d}$) and ^{10}Be ($T_{1/2}=1.5 \times 10^6\text{y}$) in the atmosphere at Tokyo and Hachijo-Island were investigated during the period of 2002 to 2003. Seasonal variations of ^7Be and ^{10}Be at Tokyo and Hachijo-Island were similar to each other. The high concentration peaks were observed in April to June and October to November. The low concentrations were observed in July to August. The isotopic ratio of $^{10}\text{Be}/^7\text{Be}$ had a peak in April to June. Using Al concentration in the aerosols, the enrichment of ^{10}Be concentration by re-suspended soil

contamination was estimated. When a box model calculation was applied to the seasonal variation of corrected $^{10}\text{Be}/^7\text{Be}$, the mean residence time in troposphere and stratosphere were 35 days and 2 years, respectively.

1A14 : ^{137}Cs inventories in the water column collected from the Sulu and South China Seas

Yamada, M., Wang, Z.-L., Zheng, J. (Natl. Inst. Radiol. Sci.)

Seawater samples were collected in the Sulu and South China Seas and their ^{137}Cs activities were determined by γ spectrometry. A significant difference in intermediate water ^{137}Cs activities in the 750-1500 m depth was observed between the Sulu and South China Seas. The ^{137}Cs inventories in the Sulu Sea was 5.7 times higher than that of the integrated deposition density of atmospheric global fallout at the same latitude of 0-10°N. A possible mechanism controlling this extremely high ^{137}Cs inventories may be inflows of ^{137}Cs rich North Pacific Tropical Water and upper North Pacific Intermediate Water through the Luzon Strait from the West Philippine Sea, and lateral transport across the Mindoro Strait into the Sulu Sea, then conveyance into the deep layer in the Sulu Sea basin.

1B01 : New phenomena - Abnormal enhancement of radon emanation by molybdophosphate complex -

Komura, K., Murata, Y., Tanaka, K., Nakamo, Y. (LLRL, Kanazawa Univ.)

Strange phenomena was found that gamma ray counts of ^{214}Pb and ^{214}Bi became much lower than expected values for the standard source prepared from ammonium molybdophosphate (AMP) and uranium (NBL 42-1) in radioactive equilibrium state. The decrease began immediately after the preparation of the source by mixing and packing in polyethylene sheet (PE). Maximum decrease was $\sim 2/3$ at room temperature and $\sim 1/4$ at 60 °C. At first this phenomena was considered to be caused by unknown (atomic-scale) interaction between AMP and gaseous radon since the large decrease up to 1/4 cannot be explained by the escape of gaseous radon through PE. Sodium molybdophosphate (SMP) showed unbelievable decrease, i.e. $\sim 1/10$ at room temperature and $\sim 1/40$ at 80 °C. Recent experiments using EVOH (Ethylene vinyl alcohol copolymer), which is known to impermeable material, suggested that the decrease of gamma ray count was caused by the escape of radon through PE. Nevertheless, it remains still mystery why 97% of radon can penetrate through the PE.

1B02 : Correlation between fission fragments and α -particles in the $^{18}\text{O} + ^{244}\text{Pu}$ reaction

Nishinaka, I.,¹ Tanikawa, M.,² Nagame, Y.,¹ Asai, M.,¹ Tsukada, K.,¹ Toyoshima, A.,¹ Ichikawa, T.³ (¹JAEA, ²Univ. of Tokyo, ³RIKEN)

The excitation energy dependence of competition of symmetric and asymmetric mass divisions of ^{258}Fm could be studied via the incomplete fusion-fission reaction of $^{244}\text{Pu}(^{18}\text{O}, \alpha)^{258}\text{Fm}$. The process of fission accompanied by α -particles in heavy ion reactions is not yet understood. In this work, correlation between fission fragments and

a-particles has been studied experimentally. Probabilities of the $^{244}\text{Pu}(^{18}\text{O}, \alpha)^{258}\text{Fm}$ reaction have been obtained from the a-particle spectra measured in coincidence with fission fragments, comparing with statistical calculations and the Q-value of the $^{244}\text{Pu}(^{18}\text{O}, \alpha)^{258}\text{Fm}$ reaction.

1B03 : Development of a novel heavy element chemistry apparatus using the RIKEN gas-filled recoil ion separator GARIS as a pre-separator

Haba, H.,¹ Kaji, D.,¹ Takabe, T.,² Akiyama, T.,³ Morimoto, K.,¹ Yoneda, A.,¹ Morita, K.¹ (¹RIKEN, ²Osaka Univ., ³Saitama Univ.)

The gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator GSRIS has been developed for the superheavy element chemistry at the RIKEN Linear Accelerator Facility. The performance of the present system was investigated using ^{206}Fr and ^{245}Fm produced in the ^{40}Ar -induced reactions on ^{169}Tm and ^{208}Pb targets, respectively. The α particles of ^{206}Fr and ^{245}Fm separated with GARIS and transported by the gas-jet were measured with a rotating wheel system for measurement of α and spontaneous fission decays. The α lines of ^{206}Fr and ^{245}Fm were clearly identified under desired low background conditions. The high gas-jet yields over 90% are independent of the beam intensities studied up to 2 pμA.

1B04 : Development of a gas-jet transport system coupled to GARIS for heavy element chemistry

Kikunaga, H.,¹ Haba, H.,¹ Kaji, D.,¹ Sato, N.,¹ Akiyama, T.,¹ Morimoto, K.,¹ Yoneda, A.,¹ Morita, K.,¹ Takabe, T.,² Ooe, K.,² Shinohara, A.,² Suzuki, D.,³ Nanri, T.,³ Yamazaki, I.,³ Yokoyama, A.,⁴ (¹RIKEN, ²Grad. School Sci., Osaka Univ.,³ Fac. Sci., Kanazawa Univ., ⁴Grad. School Nat. Sci. Tech., Kanazawa Univ.)

We plan to study the chemical properties of superheavy elements (SHEs) by developing the gas-jet transport system coupled to the gas-filled recoil ion separator (GARIS) at the RIKEN Linear Accelerator Facility (RILAC). The greatest advantage in this system is that the newly devised method makes it possible to introduce only desired nuclides for chemical studies to chemistry apparatuses. In order to efficiently transport the SHE nuclides from the target to the chemistry apparatus, it is necessary to accumulate setting parameters of GARIS and the gas-jet transport system. For this purpose, we investigated the performance of the GARIS-Gas-jet system using ^{255}No produced in the $^{238}\text{U}(^{22}\text{Ne}, 5n)$ reaction. The transport efficiency of ^{255}No to the focal plane window of GARIS was evaluated to be 6% at the optimized magnetic field. The high gas-jet yields over 80% were obtained with a rotating wheel system for measurement of α and spontaneous fission decays.

1B05 : Cation-exchange behavior of Rf in HNO_3/HF mixed solution (II)

Ishii Y.,¹ Tome H.,¹ Toyoshima A.,¹ Asai M.,¹ Nishinaka I.,¹ Tsukada K.,¹ Nagame Y.,¹ Miyashita S.,² Mori T.,² Suganuma H.,² Tashiro Y.,³ Shinohara A.,³ Sakamaki M.,⁴ Goto S.,⁴ Kudo H.,⁴ Haba H.,⁵ Akiyama K.,⁶ Oura Y.,⁷ (¹JAEA, ²Shizuoka Univ., ³Osaka Univ., ⁴Niigata Univ., ⁵RIKEN, ⁶Univ. Tsukuba, ⁷Tokyo Metropolitan Univ.)

We have investigated cation-exchange behavior of Rf and the homologues in HNO_3/HF solution using Automated Ion exchange separation apparatus coupled with the Detection system for Alpha spectroscopy (AIDA). The K_d values of Zr and Hf in 0.1 M $\text{HNO}_3/10^{-4} - 10^{-2}$ HF were decreased with increasing the concentration of the fluoride ion $[\text{F}^-]$, indicating the formation of the fluoride complexes. The K_d values of Rf in 0.1 M $\text{HNO}_3/0.005 - 0.1$ HF were also decreased with increasing $[\text{F}^-]$. It was found that there is about one-order of magnitude difference in $[\text{F}^-]$ between Rf and the homologues. This means that the strength of the fluoride complexation of Rf is much weaker than those of Zr and Hf. In the symposium, we will also present the cation-exchange behavior of Rf, Zr and Hf depending on the concentration of the hydrogen ion $[\text{H}^+]$ at the constant $[\text{F}^-]$.

1B06 : Solvent extraction behavior and electrolytic redox reaction of Tungsten under extremely low concentration

Ooe, K.,¹ Kuribayashi, T.,¹ Takabe, T.,¹ Tashiro, Y.,¹ Kitamoto, Y.,¹ Saika, D.,¹ Matsuo, K.,¹ Takahashi, N.,¹ Yoshimura, T.,¹ Takamiya, K.,² Shibata, S.,² Haba, H.,³ Enomoto, S.,³ Shinohara, A.,¹ (¹Osaka Univ., ²KUR, ³RIKEN)

Seaborgium (element 106, Sg), that is 6-group element, is homologous element of Tungsten (W). The redox reaction of Sg is expected to be observed because W has various oxidation states. For studying the chemical properties of Sg, it is important to examine chemical properties of homologous element, W. Tungsten forms polyoxometalate under high concentration, but Sg cannot form polyoxometalate because its production rate is one atom per several hours. So the chemical behavior of Sg will be different from that of W under high concentration. We investigated solvent extraction behavior and electrolytic redox reaction of Tungsten under extremely low concentration using short-lived radionuclides produced by an accelerator. From the results, it was revealed that the solvent extraction behavior of W under extremely low concentration is different from the one under high concentration.

1B07 : Application of mass spectrometry for the identification of chemical species in gas phase chemistry

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

We have studied the gas phase chemistry for transactinide elements by use of dipivaloylmethane (dpm) complex of fission products from ^{252}Cf . However, the chemical species of the generated volatile compounds were not clear. In order to argue the chemical property, it is necessary to clarify chemical species of volatile compounds. For the identification of chemical species at the gas phase chemistry, mass spectrometry with laser ionization is considered to be suitable. The resonance enhanced multi-photon ionization (REMPI) by a tunable laser is used as an ionization method.

1B08 : Studies on hot atom chemical behavior of energetic ions in solids (IX) - Effects of helium ion implantation on chemical behavior of deuterium implanted into silicon carbide -

Ishikawa, H.¹, Oya, Y.¹, Suda, T.¹, Nishikawa, Y.¹, Miyauchi, H.¹, Tanaka, S.², Okuno, K.¹ (¹Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ., ²Grad.School of Eng., The Univ. of Tokyo)

In blanket systems for future D-T fusion reactors, energetic helium and tritium are produced by nuclear reaction in tritium breeding materials and implanted into blanket structural materials. Silicon carbide (SiC) is a candidate for blanket structural materials due to its low activation. Therefore, it is important to understand chemical states and retention behaviors of tritium in SiC from a view point of fusion safety. It was previously reported that the deuterium implanted into SiC was trapped with forming Si-D and C-D bonds. In this study, helium ions were pre-implanted and then deuterium ions were implanted into SiC. Thereafter the retention of deuterium in SiC was evaluated by means of XPS and TDS and compared to that obtained for SiC implanted only with deuterium ions. These results indicate that the helium ions pre-implantation would introduced carbon vacancies and could decreased retention of deuterium with forming Si-D bond.

1B09 : Studies on hot atom chemical behavior of energetic ions in solids (X) - Ion fluence dependence on chemical behavior of energetic deuterium implanted into oxygen-contained boron film -

Kikuchi, Y.¹, Yoshikawa, A.¹, Miyauchi, H.¹, Oya, Y.¹, Sagara, A.², Noda, N.², Okuno, K.¹ (¹Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ., ²National Inst. for Fusion Sci.)

In D-T fusion reactors, boronization is expected to diminish the impurities in D-T plasma. Boron could trap impurities well, especially oxygen, and form oxides. Energetic hydrogen isotopes and helium were implanted into such boron films during fusion operations. Therefore, it is important to reveal the tritium behavior in oxygen-contained boron film from the viewpoint of fusion safety. In this study, energetic deuterium ions were implanted into 20% oxygen-contained films with the ion fluence between 0.5×10^{21} and $1.0 \times 10^{22} \text{ D}^+ \text{ m}^{-2}$, and the deuterium retention and the chemical state of trapped deuterium were evaluated by TDS and XPS, respectively. Additional experiments for pure boron films were performed under the same condition to compare with the results of oxygen-contained boron film. It was suggested that the O-D bonds were preferentially formed in boron films at lower ion fluence, and above $5.5 \times 10^{21} \text{ D}^+ \text{ m}^{-2}$, O-D bonds were saturated.

1B10 : Studies on hot atom chemical behavior of energetic ions in solids (XI) - Effects of implantation temperatures on chemical behavior of deuterium in highly oriented pyrolytic graphite -

Suda, T., Miyauchi, H., Yoshikawa, A., Oya, Y., Okuno, K. (Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ.)

In fusion reactors, carbon would be employed as plasma facing materials. It, therefore, is important from the viewpoint of more realistic estimation of tritium inventory to accumulate data for implantation temperature dependence of the T trapping states in graphite. In this study, dependence of

implantation temperatures on chemical behavior of energetic deuterium implanted into HOPG was studied by means of XPS and TDS. The experimental results indicated that the deuterium retention decreased with increasing the implantation temperature. It was found that D implanted into HOPG was trapped by two chemical states namely Peak 1 at 900 K and Peak 2 at 1050 K. They were attributed to the desorption of deuterium from sp^3 hybrid orbital type C-D bond and σ type C-D bond of sp^2 hybrid carbon, respectively. It was also found that the decrease of deuterium retention resulted in Peak 1 showed no significant dependence on the implantation temperature, while that of Peak 2 depended on the implantation temperature.

1B11 : A remote valency control technique for actinides by external ultrasound irradiation

Toraishi, T., Kimura, T., Arisaka, M. (JAEA)

We here report the reduction of Np(V) to Np(IV) induced by external ultrasound irradiation to demonstrate the ability of ultrasound for the valency control of actinide elements. High power ultrasound irradiation to water produces cavitation bubbles. The violent collapse of the bubbles generates "hot spot" with an instantaneous temperature of thousands degrees and a pressure of thousands atmosphere, and produces $\text{H}\cdot$ and $\text{OH}\cdot$ as a result. Therefore, with scavenging $\text{H}\cdot$ or $\text{OH}\cdot$, ultrasound irradiation provides either oxidizability or reducibility. 2 ml Np(V) solution in 1 M HCl was irradiated by 600 kHz external ultrasound. 10 vol% of 2-propanol and a Pt black catalyst were added to the test solution. The test solution was kept at 293 K, and bubbled by Ar gas saturated by 2-propanol during irradiation. The reduction was confirmed from the growth of characteristic light absorption of Np(IV) at 725 nm and 960 nm, together with the disappearance of the absorption of Np(V) at 980 nm. Finally, the reduction of Np(V) was completed after 180 min irradiation.

1B12 : Study of actinide complexation by calorimetry

Kirishima, A., Onishi, Y., Sato, N., Tochiyama O. (IMRAM, Tohoku Univ.)

The thermodynamics of complex formation of U(VI) and Eu(III) with mono-carboxylic and di-carboxylic acids was studied. The comparison of the thermodynamic quantities, i.e., free energy, enthalpy and entropy changes determined by potentiometric and calorimetric titration indicates that these reactions are mainly driven by the entropy change while the enthalpy change is nearly zero or preventing the progress of the reaction. This "entropy driven" character is a typical tendency of the reaction between Lewis hard acid and hard base. The result is interpreted as the bond energy "spent" for the dissociation of hydration water from the metal ion is nearly equal to the energy "earned" for the formation of new bond between metal and ligand, and these released water molecules which are now free particles increase entropy of the system dominating the driving force of these reactions. It is also discussed what kind of metal ion's character controls the entropy change.

1B13 : Separation of trivalent actinide from lanthanide by a solvent extraction technique

using imidazoledithiocarboxylic acid

Miyashita, S.,¹ Satoh, I.,² Yanaga, M.,¹ Okuno, K.,¹ Suganuma, H.¹ (¹Radiochem. Res. Lab., Fac. Sci., Shizuoka Univ., ²IMR., Tohoku Univ.)

The extraction behavior of ²⁴¹Am and ^{152,154}Eu by a solvent extraction technique using imidazoledithiocarboxylic acid (IMD) were investigated. Although the solubility of IMD into organic solvent is very poor, it was improved by the formation of ion pair with hydrophobic cation, such as tetrabutylammonium ion (TBA⁺) or tetraoctylammonium ion (TOA⁺). The obtained tetrabutylammonium imidazoledithiocarboxylate (TBA⁺IMD⁻) and tetraoctylammonium imidazoledithiocarboxylate (TOA⁺IMD⁻) are able to solve into various organic solvents, for example cyclohexanone, chloroform and nitrobenzene, but not to solve into nonpolar alkane. The radionuclides of Am(III) and Eu(III) are able to be extracted in the region of $2 < \text{pH}_{\text{eq}} < 7$ by TBA⁺IMD⁻/cyclohexanone and TOA⁺IMD⁻/cyclohexanone. The distribution ratio of Am(III) is higher than that of Eu(III) when the organic phase is 0.1 M TBA⁺IMD⁻/cyclohexanone and the aqueous phase is 1.0 M (H₂Na)NO₃. The separation factor (Am(III)/Eu(III)) at $\text{pH}_{\text{eq}} = 5.5$ is ca. 30. In the region of $\text{pH} > 6$, the distribution ratios of Am(III) and Eu(III) in the system described above showed constant values, respectively.

1B14 : Resonant X-ray emission study on electronic structure of Cm compounds

Yaita, T.¹, Shuh, D. K.² (¹JAEA, ²BNL)

Resonant inelastic x-ray scattering (RIXS), x-ray emission and absorption spectra (XES, XAS) were measured using soft x-ray based on the synchrotron radiation to determine electronic structure of Cm compounds. The partial density of state (PDOS) of N-2p of Cm-1,10-phenanthroline (Phen) complex at -5eV from Fermi level showed a significant increase and the PDOS of Eu-Phen complex, however, completely agreed with that of free phen. Accordingly, a part of chemical bond in the Cm-Phen complex includes covalent bond, while Eu-Phen complex forms through only ionic interaction. In this talk, we will present characterization of f electron behaviour on chemical bonding using RIXS.

2A01 : Variation in the atmospheric concentrations of radioactive lead isotopes with varying meteorological conditions at Kawasaki

Matsuzawa, N.,¹ Koike, Y.,² Sato, J.³ (¹Yokohama Nat. Univ., ²Univ. Tokyo, ³Meiji Univ.)

Every four hours variations of ²¹²Pb and ²¹⁴Pb concentrations were observed at Kawasaki, Japan. The variation patterns within 24 hours of the atmospheric concentration of short-lived ²¹⁴Pb is similar to that of ²¹²Pb concentration. Temporal reductions of atmospheric ²¹²Pb concentration by rainfalls also appeared. The atmospheric ²¹²Pb and ²¹⁴Pb concentrations decreased increasing wind velocity, implying that the possible turbulence in the lower layer of the atmosphere dilutes the surface air of high concentration with the air at higher layer of lower concentration. The variation in atmospheric concentrations of the short-lived ²¹²Pb and ²¹⁴Pb, which is formed within the surface air, depends strongly on the local meteorological

conditions in a short period.

2A02 : Development of the uranium-enrichment screening method for particles by chemical etching of fission track detector

Lee, C. G., Iguchi, K., Suzuki, D., Esaka, F., Magara, M., Sakurai, S., Usuda, S. (JAEA)

Safeguards environmental sample analysis has implications on the examination of the existence of uranium particles with a higher ²³⁵U enrichment in swipe samples. If such uranium particles can be preferentially detected in a sample, this will be significant in simplifying the particle analysis in the safeguards environmental samples. This method is particularly effective when analysis results are required immediately. In this study, a screening method for uranium particles according to their enrichment has been developed by using the characteristics of fission tracks, which depend on the enrichment of the uranium particles. A two-step filtration system that can collect particles with desired diameters was used for the collection of uranium particles from a swipe sample in order to avoid the influence of the differences in the diameter on the etching behavior of fission tracks. It was shown that the enrichment-based screening of the uranium particles is possible by controlling the etching time to detect fission tracks and by comparing fission track morphologies.

2A03 : Development of organically bound tritium analysis method with noble gas mass spectrometry

Kakiuchi, H.,¹ Hasegawa, H.,¹ Ichinohe, T.,² Hisamatsu, S.¹ (¹Inst. for Environ. Sci., ²Environ. Res. Center)

Tritium in biological samples exists as free water tritium and organically bound tritium (OBT). Since those chemical forms have different behavior in the environment, they need to be separately determined. The OBT is generally analyzed by measurement of ³H radioactivity by a liquid scintillation counter (LSC) after combustion of lyophilized sample. However, recent decline of OBT concentration in the general environment makes the measurement with the LSC difficult. We developed an analytical method for OBT in environmental samples by measuring triogenic ³He with a noble gas mass spectrometer, and successfully applied to low-level OBT in samples collected from the current general environment. Typical lower detection limit of this method was 0.02 Bq L⁻¹ combustion water, and an order of magnitude lower than that with the conventional method. This work was supported by contract with Aomori prefecture, Japan.

2A04 : Energy distributions of environmental neutron in liquids and solids

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

Depth profiles of environmental neutron fluxes in fresh water, seawater, concrete, iron and lead have been measured by the activation method of gold foils. Eleven targets of 16 to 33 g gold foils (40 x 300 x 0.1 mm) were irradiated for roughly 20 days. The photo peak of Au-198 was measured for 4 - 5 days by Ge detectors in the OUL at the same time. The neutron fluxes in fresh water and seawater were decreased exponentially, while those in iron and lead were not decreased

with depth deeper than 17 g cm^{-2} . A maximum has not been found in shallow depth. That in concrete had a maximum at 30 g cm^{-2} and then decreases exponentially. Depth profiles of gold activity and neutron flux were simulated, by using two Monte Carlo neutron transport codes (MCNP4C and MCNPX240) and the results were compared. The simulated depth profile of gold was almost the same as it of a thermal neutron.

2A05 : The KENS shielding experiment for high energy neutrons

Matsumura, H.,¹ Nakao, N.,¹ Masumoto, K.,¹ Oishi, K.,² Kawai, M.,¹ Aze, T.,³ Toyoda, A.,¹ Numajiri, M.,¹ Takahashi, K.,¹ Fujimura, M.,⁴ Wang, Q. B.,⁵ Bessho, K.,¹ Sanami, T.¹ (¹KEK, ²Shimizu Co., ³The Univ. of Tokyo, ⁴Nihon Univ., ⁵IHEP)

In order to obtain experimental data for benchmark of particle transport Monte Carlo codes, we performed a shielding experiment with high-energy neutrons produced from a tungsten target bombarded with 500-MeV protons and penetrated through a concrete shield in the 0 degree direction. Using an activation method, we observed many radioactive products induced by the neutrons with energies ranging from thermal to 500 MeV and obtained their production yields from various target elements at depths of 0 to 4 m from the surface of the concrete shield. We could see an attenuation of the neutrons indirectly from the variation of the production yields.

2A06 : Characteristics of Be-7 and C-11 in the cooling water of high-energy proton accelerators

Bessho, K., Chida, A., Matsumura, H., Hozumi, K., Miura, T., Masumoto, K. (KEK)

Characteristics of Be-7 and C-11 induced in the cooling water of high-energy proton accelerators were investigated. Gamma-ray measurements of the ion-exchange resin installed in the water circuit clarified that C-11 is a major gamma-emitting nuclides at the accelerator operation and is concentrated on the surface layer of the ion-exchange resin. After the beam-stop, gamma-ray activity corresponding to C-11 decreased; consequently Be-7 became major nuclides. Ion-exchange and colloid-formation of Be-7 and C-11 were examined using cationic/anionic exchange column and ultrafiltration membrane. These experiments clarified that 40% of Be-7 were dissolved in cationic form which adsorbed on the cationic-exchange resin, and the remaining 60% existed as colloid particles of which diameter is in the range of 3-7 nm. The colloids including Be-7 were also retained on the cationic-exchange resin by weak electrostatic interactions originating from the surface-charge of the particles. On the other hand, part of C-11 existed as anionic form, and 30% of C-11 existed as colloid particles. These characteristics of Be-7 and C-11 will be useful for radiation safety at high-energy and high-intensity accelerator facilities.

2A07 : Effect of oxygen on the CH_3T and CH_4 emissions from stainless steel surface

Higaki, S.,¹ Oya, Y.,^{1,2} Makide, Y.¹ (¹Radioisotope Center, The Univ. of Tokyo, ²Radiochem. Res. Lab., Shizuoka Univ.)

Unknown methane contamination occurs by the

metal-metal collision or friction of stainless steel (SS). After T_2 gas was absorbed into the inner surface and many small SS balls placed in a canister made of SS, the contamination was simulated by shaking the canister. We reported previously that methane (CH_3T and CH_4) and hydrogen (HT) were generated when helium was introduced into the canister, while neither methane nor HT was generated when only O_2 was introduced into the canister. In this presentation, we introduced helium or helium containing 10% O_2 into the canister, and investigated the emissions of CH_3T , CH_4 and HT. Under the atmosphere of helium with 10% O_2 , methane and hydrogen were not observed. It was suggested that the unknown methane contamination does not occur to the atmospheric samples stored in SS canisters, whereas occurs with SS frictions under the vacuum or without oxygen. T and/or H passed through the oxide layer on the SS surface may readily combine with O_2 than with each other or with hydrocarbons emitted from SS.

2A08 : Synthesis of molecular hexatechnetium clusters by means of dimensional reduction of their polymeric complexes

Ikai, T.,¹ Yoshimura, T.,¹ Takayama, T.,² Sekine, T.,³ Shinohara, A.¹ (¹Osaka Univ., ²Daido Inst. of Tech., ³Tohoku Univ.)

Selenide capping hexatechnetium cluster complex $[\text{Tc}_6(\mu_3\text{-Se})_8\text{CN}_6]^{4-}$ (**1**) was prepared by the reactions of one-dimensional polymer complex $[\text{Tc}_6(\mu_3\text{-Se})_8\text{Br}_4]^{2-}$ and cyanides at high temperature. Similar reaction of sulfide capping hexatechnetium cluster complex, $[\text{Tc}_6(\mu_3\text{-S})_8\text{Br}_4]^{4-}$ with cyanide gave the terminal substituted complex $[\text{Tc}_6(\mu_3\text{-S})_8\text{CN}_6]^{4-}$ (**2**). The single-crystal X-ray analysis of **1** and **2**, showed that the Tc-Tc bond lengths become longer with larger ionic radius of the face capping ligands in the order $\text{S} < \text{Se}$, while the Tc-Tc-Tc angles of **1** and **2** are 60.00° or 90.00° . IR absorption spectrum of **1** showed an absorption peak assignable to the $\text{C}\equiv\text{N}$ stretching vibration at 2103 cm^{-1} , and that of **2** showed it at 2119 cm^{-1} . Each of cyclic voltammogram of **1** and **2** showed a reversible one electron redox wave assignable to the $\text{Tc}^{\text{III}}_6 / \text{Tc}^{\text{III}}_5\text{Tc}^{\text{IV}}$ process. These redox potentials shift to the positive about 0.4V compared to those of the Re cluster analogs.

2A09 : Relationship between inhibiting effects on lipid peroxidation induced by tritiated water and change of shape of lipid membrane due to tea catechins

Kubota, M., Haga, H., Takeuchi, Y., Oya, Y., Okuno, K. (Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ.)

The influence of β -ray from tritiated water to lipid membrane and the inhibiting effect of tea catechins on lipid peroxidation have been studied. In our previous study, the inhibiting effect of (-)-epigallocatechin gallate (EGCg) which is one of tea catechins increased with increasing of its concentration below $5 \times 10^{-5} \text{ M}$, but decreased above this concentration. The interaction of EGCg with lipid membrane was considered to be a cause of decrease in the inhibiting effect. In this present study, to make clear the reason why inhibiting effect of EGCg decreased above $5 \times 10^{-5} \text{ M}$, we noted the effect of EGCg on interior of lipid bilayer using the spin probe (16-NS) method. From these results, 16-NS location was changed from lipid phase to water phase above $[\text{EGCg}] = 1 \times 10^{-3} \text{ M}$. This suggests that lipid membrane was

broken by aggregation of EGCg at the higher concentration and 16-NS went out of liposome to water, which facilitates the scavenging of 16-NS by EGCg.

2A10 : Biodistribution of ^{224}Ra and ^{223}Ra and retention of its progeny (2)

Washiyama, K.,¹ Mitsugashira, T.,² Amano, R.¹ (¹Grad. School Med. Sci., Kanazawa Univ., ²The Oarai-branch, Inst. Materials Res., Tohoku Univ.)

An alpha-particle emitter ^{223}Ra ($T_{1/2} = 11.435$ d) has been shown to be useful for the treatment of bone metastases. It emits 4 alpha-particles with energy of 26.6 MeV decaying to stable ^{207}Pb . Our previous work showed high ^{223}Ra uptake and high ^{211}Pb retention in bone compare to those of ^{224}Ra ($T_{1/2} = 3.66$ d) and its daughter ^{212}Pb . In this work the biodistribution of ^{223}Ra and the retention of its progeny in several tissues were evaluated. We also measured the biodistribution of ^{224}Ra and its daughter ^{212}Pb retention on several tissues to compare the effect of the half-life of noble gas element Rn in both decay series. Each Ra isotope was injected into different ICR, 7-week-old, male mice and dissected at 3 days after injection. The femur, blood, liver, kidney, lung, and spleen were excised and subjected to γ -ray spectrometry. The retentions of ^{211}Pb on several tissues and of ^{212}Pb were determined. The difference in retention between two decay series Pb will be discussed at the symposium.

2B01 : Electron arrangement of pionic atoms by measuring electronic KX-ray energies

Ninomiya, K.,¹ Sugiura, H.,¹ Nakatsuka, T.,¹ Itsuki, Y.,¹ Nakashima, K.,¹ Ikai, T.,¹ Sato, W.,¹ Yoshimura, T.,¹ Matsumura, H.,² Miura, T.,² and Shinohara, A.¹ (¹Grad. School Sci., Osaka Univ., ²KEK)

Although the formation process of pionic atoms has been studied well, the electron rearrangement process after the pion capture has not been studied sufficiently. The electronic X-ray energies of pionic atoms are influenced by the atomic pions and electron arrangement. Our group has been studying on the electron structure for the pionic atoms by measuring electronic X-ray energies. All measurements were performed at 12GeV proton synchrotron of High Energy Accelerator Research Organization. Various metals and compounds ranging from $Z=29$ to 92 were used as target. In this paper, we will discuss the electron arrangement after pionic atom formation by using theoretical approach.

2B02 : Nuclear relaxation and electronic state of ^{140}Ce implanted in highly oriented pyrolytic graphite

Sato, W.,^{1,2} Ueno, H.,² Taniguchi, A.,³ Itsuki, Y.,¹ Kasamatsu, Y.,⁴ Shinohara, A.,¹ Asahi, K.,^{2,5} Asai, K.,⁶ Ohkubo, Y.³ (¹Osaka Univ., ²RIKEN, ³Kyoto Univ., ⁴JAEA, ⁵Tokyo Inst. Tech., ⁶Univ. Electro-Commun.)

The TDPAC technique has been applied to studies of the dynamic and electronic properties of Ce ions as extremely dilute impurities in an HOPG sheet. From the temperature-dependent exponential relaxation of the directional anisotropy of the angular correlations, a thermally activated dynamic motion of the probe nucleus relative to the

extranuclear charge distribution was suggested. The activation energy of the dynamic motion was estimated to be $E_a = 0.029(3)$ eV. As regards the electronic property of the probe, the chemical state of the component in motion was found to be trivalent, Ce^{3+} , by the deduced value of the paramagnetic correction factor by a TDPAC measurement under an external magnetic field. The temperature-independent second component possibly corresponds to those occupying sites damaged by the heavy-ion implantation.

2B03 : Mössbauer spectroscopy and nuclear resonant forward scattering of $\text{RRu}_4\text{P}_{12}$ (R=rare earth metals)

Tsutsui, S.,¹ Kobayashi, Y.,² Okada T.,² Haba H.,² Onodera H.,³ Yoda, Y.,¹ Sekine, C.,⁴ Shirotani, I.,⁴ Kikuchi, D.,⁵ Sugawara, H.,⁶ Sato H.,⁵ (¹JASRI, ²RIKEN, ³Tohoku Univ., ⁴Muroran Inst. Tech., ⁵Tokyo Metro. Univ., ⁶Tokushima Univ.)

We have performed ^{149}Sm nuclear resonant forward scattering and ^{99}Ru Mössbauer spectroscopy of $\text{RRu}_4\text{P}_{12}$ (R=La, Pr, and Sm). In the sample of $\text{SmRu}_4\text{P}_{12}$, the quantum beats of ^{149}Sm nuclear resonant forward scattering shows there exists a magnetic order below 4.5 K. Temperature dependence of time-integrated intensity in ^{149}Sm nuclear forward scattering suggests that magnetic ordering occurs just below metal-insulator transition temperature. No significant changes of ^{99}Ru Mössbauer parameters are observed from 5 K up to 77 K in these spectra. These results indicate that magnetic hyperfine interactions observed in $\text{SmRu}_4\text{P}_{12}$ up to now is mainly caused by magnetic dipole moments at Sm sites, originally ascribing the magnetic ordering in NpO_2 . We will also discuss the electronic and vibration states at Ru atoms in $\text{LaRu}_4\text{P}_{12}$ and $\text{PrRu}_4\text{P}_{12}$.

2B04 : Mössbauer spectroscopic studies of alkylammonium iron(III) complexes

Katada, M., Kozawa S., Nakajima Y. (Tokyo Metro. Univ.)

Alkylammonium iron(III) complexes, $[(n\text{-C}_n\text{H}_{2n+1})\text{mNH}_4\text{m}]_3[\text{Fe}(\text{CN})_6]$ were prepared and studied by Mössbauer spectroscopy, XRD, and DSC. In the complexes with $m=2$, the temperature dependences of the area intensity of Mössbauer are correlated to the motion of alkyl chains. The temperature dependence of the complex with $n=4$ was linear and smaller than that of other complexes. Especially in the complex with $n=6$, the deviation from the linear was the largest in the complexes observed. This result is attributed to the structural difference of the complex. The complexes with $n \geq 8$ consist of two-dimensional layer structure. The temperature dependence of the area intensity was similar to each other. This means that the motion of alkyl chain in these complexes are almost the same. The values of quadrupole splitting for the complexes were larger those that of the complexes ($m=1$). This indicates that the form of $[\text{Fe}(\text{CN})_6]^{3-}$ ion is affected by the differences of the number of alkyl groups.

2B05 : Mössbauer studies and magnetic susceptibility of SnO_2 doped with ^{57}Fe

Sakuma J.,³ Bareero, Cesar. A.,^{1,2} Nomura K.,¹ and Takeda M.,³ (¹The Univ. of Tokyo, ²Univ. de Antioquia, Colombia., ³Toho

Univ.)

SnO₂ powders doped with various ⁵⁷Fe contents were prepared by a sol-gel method and characterized by Mössbauer spectroscopy, vibrating sample magnetometer (VSM), and X-ray diffraction (XRD). The results suggest that our samples consist only of defective rutile type SnO₂, which are characterized by having tin interstitial, high spin Fe³⁺ in octahedral coordination and oxygen vacancies. The samples seem to exhibit many sources and types of magnetism, and the dominance of one of them greatly depends upon the synthesis conditions. The samples seem to exhibit many sources and types of magnetism, and the dominance of one of them greatly depends upon the synthesis conditions. One sample exhibits ferromagnetism and shows a relaxed Mössbauer sextet, suggesting that the main source of magnetism seems to be related with ordered iron ions. On the other hand, in another sample we observed ferromagnetism but its Mössbauer spectrum consists only of doublets, thus the main source seems to be related with defects. Finally, another sample exhibits an intensive sextet with sharp lines but negligible or no magnetization, the observed magnetism is mainly attributed to the presence of hematite.

2B06 : The study on the iron(III) LIESST compounds

Hayami, S., Hiki, K., Sato, O., Maeda, Y. (Kyushu Univ.)

The LIESST effect for [Fe(pap)₂]ClO₄ (**1**) has been made to clear by investigation for the HS → LS relaxation. The activation energy and the preexponential factor with the HS → LS relaxation are $E_a = 1816 \text{ cm}^{-1}$ and $k_{HL}^0 = 4.07 \times 10^4 \text{ s}^{-1}$. It was estimated that the tunneling rate observed at low temperatures is $k_{HL}(T \rightarrow 0) = 2.8 \times 10^{-8} \text{ s}^{-1}$. The $k_{HL}(T \rightarrow 0)$ value for the iron(III) compound **1** is found to be unreasonably smaller than the rate expected for iron(III) compounds, and even smaller than the tunneling rate for many iron(II) compounds. It is thought that the LIESST effect for iron(III) compounds has been observed in order to have the strong intermolecular interactions. What kind of processes eventually lead to such long-lived metastable HS states for iron(III) compounds has remained an open question. Here we have discussed the structure, HS → LS relaxation, cooperativity about LIESST effect for iron(III) SCO compounds. It is able to suggested that the strong intermolecular interaction play role in the trapping photo-induced metastable HS state for iron(III) compounds.

2B07 : Spin-crossover phenomena of the assembled complexes and their Mössbauer spectra

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

Assembled complexes, Fe(NCX)₂(bpa)₂ (X=S, Se, BH₃; bpa=1,2-bis(4-pyridyl)ethane) show spin-crossover phenomena by enclathrating organic molecule. The ratio of spin transition and the transition temperature depended on the assembled structure (1D, 2D grid, or interpenetration structure). The cooperativity coefficients were estimated to be 0.96-1.33. The QS value of Fe(II) LS state was 0 mm s⁻¹, suggesting a good symmetry around iron atom. There was a good relation between QS of Fe(II) HS state and transition temperature, i.e., the complex having smaller QS value

shows higher transition temperature. This relation was discussed from the ligand field theory.

2B08: Mössbauer spectra of new cathode material for lithium-ion battery

Nishida, T.,¹ Yoshida, Y.,¹ Takahashi, H.,² Okada, S.,² Yamaki, J.² (¹Kinki Univ., ²Kyushu Univ.)

Mössbauer spectroscopy is very effective for the development of lithium ion battery. Mössbauer spectra of LiFeVPO_x, LiFeV_{0.5}PO_x and LiFePO_x glasses prepared by conventional melt-quenching method consist of one doublet peak due to Fe^{III} with an octahedral symmetry. Heat treatment of LiFeVPO_x and LiFeV_{0.5}PO_x glasses at around crystallization temperature caused a marked decrease in the values of Δ , reflecting a structural relaxation of the glasses accompanying a remarkable decrease in the distortion of Fe^{III}O₆ octahedra. After heat treatment, a marked increase in the electrical conductivity was observed from the order of 10⁻⁶ S cm⁻¹ to 10⁻³ S cm⁻¹. As a result of heat treatment, a remarkable increase in the specific discharge- and charge-capacity of the lithium-ion battery (coin cell) was observed from 50 to 150 mAh g⁻¹. These results prove that structural relaxation of cathode active material promotes intercalation and deintercalation of Li⁺ in the coin cell during discharge and charge processes, respectively.

2B09 : Photocontrol of the electronic states of iron coordinated to photoresponsive polyaniline.

Oshida, K., Einaga, Y. (Keio Univ.)

Recently, photofunctional materials have attracted much attention in the high density information technology. However, some limitation remain to develop applications from such materials. To overcome this issue, we have reported several optical switchable magnetic materials containing photochromic compounds. In this experiment, we have designed a novel type of hybrid composited material consisting of PANI having photoresponsive azobenzene in the main chain and ferric chloride (II) as inorganic metals. As a result, we have succeeded in controlling the electronic states of iron coordinated to PANI backbone by illumination at room temperature.

2B10: Effective application of Mössbauer spectroscopy to chemical education - Support to general chemistry laboratory of redox reactions of iron -

Sakai, Y.,¹ Takayama, T.,¹ Ogiso, R.,¹ Onaka, S.,¹ Watanabe, Y.,¹ Sano, H.² (¹Daido Inst. Tech., ²Ohtsuma Women's Univ.)

One of the educational programs of general chemistry laboratory at Daido Institute of Technology is "redox reactions of iron", which was started recently. The aim is for students to acquire a basic concept of redox via familiar phenomena. We used ⁵⁷Fe-Mössbauer spectroscopy effectively in constructing this educational program. In our program for about 3 hours, we have three parts; (1) observing the chemical reactions, which are essential through the whole program, between Fe²⁺ or Fe³⁺ aqueous solution and [Fe(CN)₆]⁴⁻ or [Fe(CN)₆]³⁻ aqueous solution, (2) the Evance's experiment showing that it is Fe²⁺ that should be firstly produced in oxidizing corrosion of metallic iron immersed in an aqueous solution, and (3) preparation of blueprints (or

cyanotypes), making students to understand light-induced reduction reactions of iron species. From our Mössbauer measurements, it was confirmed a very important key-factor that the reaction between Fe^{2+} and $[\text{Fe}(\text{CN})_6]^{3-}$ (ferricyanion) yields deeply-blue colored precipitation in both the (2) and (3) part.

3P01 : Multi-element analysis of cultivated oyster tissues from Miyagi Pref.

Fukushima, M.,¹ Nakano, Y.,² Chatt, A.³ (¹Ishinomaki Senshu Univ., ²Kyoto Univ. Reactor, ³Dalhousie Univ., SLOWPOKE-2)

Oyster tissues cultivated in six different bays in Miyagi Pref. were analyzed by neutron activation analysis (NAA) to study the influence of environment to cultivated oysters. Oysters were collected from Shizugawa, Nagatsura-Hama, Onagawa, Ishinomaki, Naruse, and Matsushima Bay in October, 2005. Soft tissues were separated into four parts; mantle, gill, muscle, and hepatopancreas. Each organ was freeze dried and pulverized. About 0.5g of dried powder was irradiated in Kyoto University Reactor, Japan, (KUR) and another portion was irradiated in SLOWPOKE-2, Dalhousie Univ., Canada (DUSR). By NAA in both facilities, Ag, Br, Ca, Cl, Co, Cr, Cu, Fe, I, Mg, Mn, Na, Rb, Sb, Se, V, and Zn were analyzed. Differences of elemental levels were found for Na, Cl, Fe, and Zn. As for Fe, rather high levels were found in oysters from Shizugawa and Matsushima Bay. As for Zn, rather low levels were found in oysters only from Nagatsura-Hama.

3P02 : A study on the selective distribution of cobalt in *Clethradease* leaves by solvent extraction with supersonic waves

Kasahara, S.,¹ Maekawa, M.,^{1,2} Noya, Y.,¹ Oguri, K.,² Ohno, S.,³ Seki, K.¹ (¹Central Inst. Isotope Scie., Hokkaido Univ., ²Aichi Pref. Univ., ³Field Sci. Center North. Bio., Hokkaido Univ.)

Clethra barbinervis is known to accumulate cobalt (Co) in anomalously high contents.¹ Previously we have successfully pictured the two-dimensional distribution of Co on an imaging-plate by the combination of solvent extractions and radio activation analysis, demonstrating that cobalt mainly accumulate in sac-like features on the rim of the leaves.² Extraction with solvents, however, involved unexpected variation in the abstraction efficiency, presumably depending on the drying conditions of the leaves. Hence, improvement of more effective extraction method is necessary to obtain more insight into the accumulation mechanism. In the present paper, we describe our findings that introduction of ultrasonic waves resulted in a great deal of shortening in extraction time, and provided an efficient extraction of contaminant metals from the leaves, with Co atoms left there. [1] N. Yamagata, S. Murata, *Koshu Eiseiin Kenkyu Hokoku* (1964) **13** (3), 170-5. [2] S. Kasahara, et al., *J. Nuclear & Radiochem. Sci.*, (2005) **6**, Suppl., 165.

3P03 : Determination of concentrations of trace elements in pancreatic cell of zinc deficiency mice

Kawashima, M., Kamishima, J., Matsushita, K., Minayoshi, R., Noguchi, M., Suganuma, H., Yanaga, M. (Fac. of Sci.,

Shizuoka Univ.)

Concentrations of various trace elements in pancreatic cell of Zn-deficient mice and control mice were determined. Eight-week old male mice were divided into Zn-deficient and control groups. After three weeks treatment, their pancreata were removed and homogenized with HEPES butter. The homogenates were separated into 6 fractions, such as nondestructive, nuclear, mitochondrial, lysosomal, microsomal, cytosolic fractions, by centrifugation under different conditions. Each fraction was freeze-dried for instrumental neutron activation analysis. Concentrations of 11 elements, Na, Mg, Cl, Mn, Fe, Co, Zn, Cu, Se, Br, Rb, in each subcellular fraction were determined by INAA. Furthermore, SDS-PAGE was carried out for the cytosolic fraction. The INAA results showed that zinc concentrations of Zn-deficient mice were lower than those of control mice and Co concentrations of Zn-deficient mice were higher than those of control mice. No significant differences were found for the positions and number of protein bands on the gel after SDS-PAGE for the cytosolic fraction.

3P04 : Variation in concentrations of trace elements and metalloproteins in hepatocyte of zinc deficient mice

Kamishima, J.,¹ Ogi, T.,¹ Minayoshi, R.,¹ Kawashima, M.,¹ Kinugawa, N.,¹ Suganuma, H.,¹ Noguchi, M.,¹ Ishikawa, K.,¹ Sera, K.,² Yanaga, M.¹ (¹Fac. of Sci., Shizuoka Univ., ²Cyclo. Res. Center, Iwate Med. Univ.)

Eight-week-old male mice of ICR strain were divided into two groups; one was fed with zinc deficient diet, and the other with control diet. After 3 weeks, their livers were homogenized. Then, the cytosolic fraction was separated by centrifugation. Two type experiments for cytosolic fraction were performed. In the first experiment, SDS-PAGE and silver stain were performed. And after gel electrophoresis, the gel was cut into some protein bands and their bands were subjected to PIXE analysis. In the second experiment, two-dimensional electrophoresis was performed. Concentrations of trace elements in each protein band were determined by PIXE analysis and zinc concentration in each band was standardized with the silver concentration, i. e., normalized with the protein amount. From this result, there are no significant differences in almost all bands between zinc deficient mice and control ones. Therefore, it is considered that some zinc binding proteins were decreased (or disappeared) under zinc deficient condition.

3P05 : Environmental evaluation of tideland sediments by neutron activation analysis and Moessbauer spectroscopy

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We collected sediments vertically in Yatsu tideland and Sanbanse tideland in order to investigate how reclamation influenced the environment of tideland sediments. Yatsu tideland was reclaimed from every direction and connected to the sea with two narrow rivers. Using instrumental neutron activation analysis and prompt gamma-ray analysis, we got vertical distribution of over thirty elements. Chemical states of iron were investigated by Moessbauer spectroscopy. As a result of measurement, there was a big change of depth

profiles in Yatsu tideland. Water content, a lot of elemental contents and chemical states of iron changed greatly in deeper section than 25cm in Yatsu tideland. On the other hand, there was not a big change in the Sanbanse tideland. It is thought that because Sanbanse tideland was located in the Tokyo bay, influence of reclamation was small. It is suggested that this trend is explained in terms of the difference between situation of reclamation in Yatsu tideland and that in Sanbanse tideland.

3P06 : Determination of extractable organohalogenes (EOX) by instrumental neutron activation analysis (INAA) and individual persistent organohalogen compounds by gas chromatography-mass spectrometry (GC-MS) in soil and sediment samples

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High amounts and many kinds of organic compounds have been produced and use in present modern society. Some of them are persistent in the environment and toxic/accumulative in humans and wildlife. Now, twelve man-made chemicals were listed up as persistent organic pollutants (POPs) and monitored in worldwide under the Stockholm Convention. However, it is worried that unknown/unidentified POPs would already pollute the environment. Analysis of organically bound halogens will be of help to understand the levels of unknown/unidentified organohalogenes when organically bound halogens and identified organohalogen concentrations are compared. In this study, organically bound halogens were determined by instrumental neutron activation analysis (INAA) in the fraction extracted with organic solvents (extractable organohalogenes: EOX). Consequently, in order to reveal the worldwide distribution of known and unknown/unidentified POPs in soil and sediment, determination of EOX and known POPs by gas chromatography-mass spectrometry (GC-MS) were conducted. The results indicated that known POP levels are normally only around less than ten per cent of EOX levels, suggesting the presence of high concentrations of unknown/unidentified organic halogen compounds in soil and sediment samples.

3P07 : Study on presolar grain using neutron activation analysis with multiple gamma detection

Hatsukawa, Y., Miyamoto, Y., Toh, Y., Oshima, M., Hayakawa, T. (JAEA)

Method of multiparameter coincidence spectrometry based on gamma-gamma coincidence is widely used in the field of nuclear structure studies, and has produced many successful achievements. We developed a new technique of trace element analysis based on neutron activation analysis with coincident gamma-ray detection. In order to improve the low efficiency of coincident gamma-ray detection, an array of 16 Ge detectors with BGO Compton suppressors was used for this method. High sensitive trace element analyses without chemical separation were carried out by the combination of neutron activation analysis and the method of multiparameter coincidence spectroscopy. In this study, we try to measure trace elements in presolar grains using neutron activation

analysis with multiple gamma ray detection. Presolar grains are synthesis in stars such as the Super nova or Red giant, and found in the primitive meteorites. We obtained 160 micro grams of nanodiamonds which is one of presolar grains from the Allende meteorite, and measured trace elements in the grains using this method. 8 elements in presolar grain were detected.

3P08 : Trace elemental abundances in seawater determined by INAA using a multiple gamma-ray detector

Takata, Y.,¹ Oura, Y.,² Ebihara, M.,² (¹Univ. of Tokyo, ²Tokyo Metropolitan Univ.)

In a lot of fields instrumental neutron activation analysis (INAA) method has been widely used to determine the concentration of trace elements since it can determine multi-elemental concentrations simultaneously. But now, INAA is not applied to seawater samples. Because main component elements of seawater such as Na, Cl, and Br produce high radioactivity after neutron irradiation, so it was nearly impossible to measure gamma-rays of short life radionuclides. To avoid the interference of the high radioactivity produced from main components, a multiple gamma-ray detector called GEMINI-II was applied to measure gamma-rays from seawater samples after neutron irradiation. Concentration of Sc in seawater, which has been hardly reported so far, was able to be determined nondestructively only with a procedure of drying.

3P09 : Evaluation of activable tracer for nuclear medicine

Washiyama, K., Amano, R. (Grad. School Med. Sci., Kanazawa Univ.)

Almost of all rare earth elements (REEs) have relatively high activation cross section and determined sensitively by neutron activation analysis (NAA). Moreover, not only determinations of the biodistribution of REEs by NAA, but also the concentration of trace metal element which may influence by REEs toxicity are also simultaneously determined. In this study, to evaluate the usefulness of activable tracer for nuclear medicine, radioactive tracer ¹⁵³SmCl₂ and activable tracer ^{nat}SmCl₂ were used and the biodistribution of samarium in mice were compared. Radioactive tracer ¹⁵³SmCl₂ was produced by irradiating ^{nat}Sm₂O₃ at Kyoto University research reactor (KUR). Male 4-week-old ICR mice (n=9) were administered ¹⁵³SmCl₂ with three different concentration of stable samarium. Mice were sacrificed and dissected 1 h after ¹⁵³SmCl₂ injection. The blood, liver, kidney, femur, brain, and muscle were excised and subjected to γ -ray spectrometry. The results were expressed as percent dose per gram (%dose/g) of tissue. In the activable tracer experiment, mice (n=9) with the same condition were administered ^{nat}SmCl₂. Mice were sacrificed and dissected 1 h after injection and the same tissues were excised. The samples were irradiated at KUR and the ^{nat}Sm uptake rate was determined.

3P10 : Stability of ¹⁸⁶Re- and ¹⁸⁸Re-DMSAs and removal of impurity in the product

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N.,² Hashimoto K.,³ Sorita T.³ (¹Meiji Univ., ²Univ. Tokyo, ³JAEA)

Complex compound of ¹⁸⁶Re and ¹⁸⁸Re with meso-2,3-dimercaptosuccinic acid (DMSA) is expected to be effective against cancerous disease. This compound is synthesized with high radiochemical yield by use of SnCl₂ as reducing reagent¹⁾. However, the product contains a large amount of SnCl₂ which is harmful to humans body. The removal of tin impurity was tested with cation exchange resin. ¹⁸⁶Re-DMSA complex was also obtained with high radiochemical yield by use of other harmless reducing agents (L-ascorbic acid, Na₂SO₃, H₃PO₃). Synthesized ¹⁸⁶Re-DMSA by use of SnCl₂ was stable in sodium acetate buffer for 5 hours, while survival amount of ¹⁸⁶Re-DMSA synthesized by other reducing agents decreased in a short time.

1) J. Singh *et al.*, *Nucl. Med. Commun.*, **14**, 197-203 (1993)

3P11 : Simultaneous production of high-pure ⁷⁶Br, ⁷⁷Br and ⁷⁹Kr using an isotope separator

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A production method of the bromine radioisotopes ⁷⁶Br and ⁷⁷Br in high radiochemical purity has been developed without using an isotopically enriched target, instead, their precursors ⁷⁶Kr and ⁷⁷Kr produced in a NaBr target by proton irradiation were mass-separated and implanted into Al foils together with ⁷⁹Kr by means of a high-resolution isotope separator. The isotope separation efficiencies of the radiokryptons with a compact ECR ion source were found to be 15±2%. The ⁷⁹Kr was recovered cryogenically from the Al foil heated, while ⁷⁶Kr and ⁷⁷Kr were allowed to decay for 14.6 h and 1.24 h, respectively during which the almost maximum growth of ⁷⁶Br and ⁷⁷Br occurred. A comparison was made between anion and cation exchange methods for chemical separation of the radiobromines from the Al foils. The recovery efficiencies were found to be almost identical between then, and better than 95%.

3P12 : Determination of the neutron flux at the self-shielded PET cyclotron of the university of Tokushima using the activation foil method

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At the beginning of the fiscal year, PET Tracer Production System for PET diagnosis had been installed in Tokushima University Hospital following Kagawa University Hospital in the Shikoku Island area, Japan. This system has the self-shielded PET cyclotron HM-12 (Sumitomo Heavy Industries, Ltd.) in the cyclotron building to synthesize a ¹⁸F positron emitter labeling the radio-compound as [¹⁸F]FDG. From a radiation safety management point of view, few studies have been carried out on the flux of thermal and fast neutrons and those leak neutron doses in the type of self-shielded cyclotron. To estimate the amount of radiation exposure received by medical co-workers and radiation activation of the body or shield material of the cyclotron, it should be necessary to measure the neutron flux and neutron dose leakages correctly. In this work, the activation foil methods of ¹⁹⁷Au(*n*, γ) ¹⁹⁸Au, ¹¹⁵In(*n*, *n'*) ^{115m}In and so on are

applicable to determine the flux of thermal and fast neutrons at various points in the self-shielded cyclotron, and we will report the distribution of neutron flux there.

3P13 : Oxidative stress by Se-deficiency and dynamics of biotrace elements

Sakuma Y., Tsuyuki S., Nagayama A., Matsuoka K., Honda C., Endo K. (Showa Pharmaceutical Univ.)

Metal ions such as Mn, Fe, Cu, Zn, Se are located at the center of the antioxidants in biological systems. Selenium (Se) is contained in GSH-Px (glutathione peroxidase), one of the antioxidants, and is related to various other metal ions to keep redox balance in organism. In the present study, Mn contents in cell fractions of liver homogenates of Se-deficient rat, and control (normal) rats were determined by means of INAA. The results showed that the Mn contents in microsomal fraction of male Se-deficient rats were greater than the control for male rats, and that the trend was different for female rats. The results were presented together with the results of SOD and TBARS.

3P14 : Study on inhibition mechanism of tea catechins on beta-ray-induced lipid peroxidation by examination shapes of liposome

Kubota, M., Kinoshita, W., Haga, H., Takeuchi, Y., Oya, Y., Okuno, K. (Radiochem. Res. Lab., Fac. of Sci., Shizuoka Univ.)

Previous studies have indicated that the inhibiting effect decreased with increasing of the concentration of (-)-epigallocatechin gallate (EGCg), which is one of tea catechins, above 5×10⁻⁵ M, and interaction of EGCg with lipid membrane would induce the decrease of inhibiting effect. In this present study, to elucidate the mechanism of decrease of the inhibiting effect, shape changes of lipid membrane by four kinds of tea catechins (EGCg, (-)-epigallocatechin (EGC), (-)-epicatechin gallate (ECg), (-)-epicatechin (EC)) were observed by absorptiometer and transmission electron microscope. These results suggested that EGCg induced aggregation of liposomes and shapes change of the membrane. In addition, the influence of hydrogen peroxide (H₂O₂) induced by autoxidation of catechin was measured by electron spin resonance to elucidate influence of autoxidation of EGCg on the decrease of inhibiting effect. This result suggested that the decrease of the inhibiting effect didn't be caused by the H₂O₂, but induced by shapes change of the lipid membrane due to EGCg.

3P15 : Activity ratio of Pm-146/Pm-147 in Pm-147 radiation source

Sato, T. (JAEA)

Activity of Pm-147 in same consumer goods (glow starter for fluorescent light) is measured by Ge- γ ray spectrometer. Activity of Pm-147 in glow starters scattered between 1.3E3 ~5.3E3 Bq. Together with Pm-147, Pm-146 was found in the glow starters. Activity ratio of Pm-146/ Pm-147 ranged from 5.8E-5 to 1.1E-4. The ratio is too high in comparison with ratio of cumulative fission yield of Pm-146/Pm147 for U-235 thermal fission, 2E-10. Calculation of the ratio of Pm-146/ Pm-147 by ORIGEN2 shows 2.47E-5 for PWR (burnup 33

GWD/MT) and 1.53×10^{-5} for BWR (burnup 27.5 GWD/MT). These ratios agree with the ratio found for glow starters taking account of decay of two nuclides Pm-146 ($t_{1/2}=5.53$ y), Pm-147 ($t_{1/2}=2.26$ y). As no cumulative fission yield of Pm-146 in the ORIGEN2 library, production path of Pm-146 is not fission but other nuclear reaction such as Pm-147(n, 2n)Pm-146.

3P16 : Study on Hf metallofullerenes using ^{181}Hf radioactive-tracer(2)

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Recently, metallofullerenes of Group 4 elements, Ti and Hf, have also been investigated. Hf metallofullerenes have already been reported for Hf_2C_{80} and HfC_{84} , and determined as $\text{Hf}_2\text{C}_2@\text{C}_{78}$ for the former species. However, the structures and characters of these species are still unknown because of their low production yield. Here, we report the HPLC elution behavior of Hf metallofullerenes ($\text{Hf}_2\text{C}_2@\text{C}_{78}$, Hf_3C_{82} , and Hf_2C_{82}) on 5PBB, Buckyprep, Buckyprep-M and Buckyclutcher columns by use of ^{181}Hf radioactive-tracer and discuss the chemical properties from the HPLC elution behavior of Hf metallofullerenes on each stationary phase. According to capacity factor k' on 5PBB column, we consider that the number of transferred electrons from metal atom to the cage, is 6 for $\text{Hf}_2\text{C}_2@\text{C}_{78}$ and 8 for Hf_3C_{82} , and Hf_2C_{82} .

3P17 : Synthesis of endohedral ^{133}Xe -fullerenol by using higher fullerene

Watanabe S., Katabuchi T., Ishioka N. S., Matsushashi S. (JAEA)

Hydrophilic endohedral ^{133}Xe -fullerenols [$^{133}\text{Xe}@\text{C}_{76}(\text{OH})_x$ and $^{133}\text{Xe}@\text{C}_{84}(\text{OH})_x$] were synthesized from hydrophobic endohedral ^{133}Xe -higher fullerenes ($^{133}\text{Xe}@\text{C}_{76}$ and $^{133}\text{Xe}@\text{C}_{84}$). The endohedral ^{133}Xe -higher fullerenes were produced by implantation of ^{133}Xe ions into fullerene targets (C_{76} and C_{84}) using an isotope separator. After implantation, the fullerene targets were dissolved in *o*-dichlorobenzene. The solutions were filtered through a membrane filter to remove insoluble materials. For synthesis of endohedral ^{133}Xe -fullerenols, endohedral ^{133}Xe -fullerenes dissolved in *o*-dichlorobenzene were shaken with tetrabutylammonium hydroxide and KOH solution for 1 min. On this shaking, endohedral ^{133}Xe -fullerenols formed in *o*-dichlorobenzene phase. The *o*-dichlorobenzene phase was separated from the KOH phase, and pure water was added to the *o*-dichlorobenzene phase. The mixture was shaken for 1 min to extract endohedral ^{133}Xe -fullerenols into pure water phase. The recovery of the $^{133}\text{Xe}@\text{C}_{76}(\text{OH})_x$ and $^{133}\text{Xe}@\text{C}_{84}(\text{OH})_x$ will be discussed in connection with reaction conditions.

3P18 : Analysis of Am-242m in solidified products

Fujiwara, A., Kameo, Y., Haraga, T., Nakashima, M. (JAEA)

When solidified products made from low-level radioactive waste with melting treatment are disposed of in near surface repositories, concentrations of the radionuclides contained in the waste package have to be evaluated. Americium-242m is one of the targeted nuclides. In this study, simple analytical

methods to evaluate the activity of ^{242m}Am are studied. Americium, Cm and Eu in a sample solution made from simulated solidified product and tracer of ^{152}Eu and ^{244}Cm and unknown solution of $^{241+242m+243}\text{Am}$ were separated from matrix elements with TRU resin. The fraction containing Eu, Am and Cm was passed through pyridine resin to separate Am from Eu and Cm. The contamination factor of $^{244}\text{Cm}/^{241}\text{Am}$ was less than 3.0×10^{-3} . Curium-242 generated from ^{242}Am was measured to evaluate the activity of ^{242m}Am . The fraction containing Eu, Am and Cm was also passed through TEVA resin to separate Am and Cm from Eu. Beta-ray of separated Am and Cm fraction was measured to evaluate the activity of ^{242m}Am . The activity of ^{242m}Am evaluated by beta-ray measurement was agreed with that by alpha-ray measurement of ^{242}Cm .

3P19 : Modeling of cesium sorption onto sedimentary rock

Doi, R., Xia, X., Shibata, M., Kitamura, A., Yoshikawa, H. (JAEA)

Assessment for radionuclides sorption onto a rock in deep underground is important for the safety assessment of HLW geological disposal. Distribution coefficient, K_d , is an available parameter to assess the sorption in the current safety assessment. We applied a model that the sorption of Cs is dominated by the ion exchange reaction on illite to experimental results of Cs sorption onto Horonobe sedimentary rock. The equilibrium concentration of Cs was calculated using geochemical calculation code, PHREEQC, under the conditions same as those adopted in the experiment to estimate the K_d value. Comparing the calculated results with the experimental data, the model could explain well the dependence of K_d values on Cs equilibrium concentration. By using the model, it is possible to assess Cs sorption onto a sedimentary rock. However, some differences were also obtained between the experimental data and the calculated one. The uncertainty of analytical techniques for illite content is considered as a possible cause.

3P20 : Separation of uranium using microcapsules with tri-n-octylphosphine oxide

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We have been developing the separation method of uranium from sulfuric acid solution used for the decontamination of uranium bearing wastes. Decreasing the uranium concentration of the waste solution will result in the reduction of waste volume to be disposed of underground. The applicability of microcapsules with tri-n-octylphosphine oxide (TOPO) was investigated for the uranium separation method. The microcapsules with TOPO were prepared by dispersing the powder of TOPO into the alginate gel. The microcapsule was equilibrated with the sulfuric acid prior to the absorption experiments. The diameter of capsules was 250-500 μm and the TOPO content was about 6wt.%. Absorption experiments were carried out using the microcapsule and the simulated waste solution containing 0.1-1 mol/L of sulfuric acid, 100 ppm of U, and 1,500 ppm of Fe. The distribution coefficient of uranium was about 100 mL/g, and separation factor of U and Fe exceeded 1,000. The microcapsule of TOPO can be used for the uranium separation from sulfuric waste solution.

3P21 : The development of expeditious and high efficient separation method of trivalent Actinides using capillary electrophoresis

Kuribayashi, T.,¹ Kitamoto, Y.,¹ Tashiro, Y.,¹ Yoshimura, T.,¹ Mitsugashira, T.,² Shinohara, A.¹ (¹Osaka Univ., ²Tohoku Univ.)

It has been recognized that separation of trivalent actinides is difficult because their chemical properties are very similar. We have been studied the rapid separation of Am, Cm and Cf by means of capillary electrophoresis (CE). The separation using α -HIBA as a ligand were examined for the various conditions such as pH, length of capillary, and inner diameter of capillary, flow rate to transport, and chemical condition at inner wall capillary. We have previously got the result that the trivalent ion radius of Am and Cm were not lined up the tendency of the lanthanides when we plotted the mobility against the trivalent ion radius. To advance the repeatability, we have been tried to upgrade the injection and the fraction method of CE. We report the automation of fraction preparative isolation and the result of separation of Am, Cm and Cf.

3P22 : Separation of trivalent actinides from lanthanides using a capillary electrophoretic technique

Mori, T.,¹ Ishii, Y.,¹ Hayashi, K.,¹ Yanaga, M.,¹ Satoh, I.,² Suganuma, H.¹ (¹Radiochem Res. Lab., Fac. Sci., Shizuoka Univ., ²IMR, Tohoku Univ.)

A separation of ^{152, 154}Eu(III) and ²⁴¹Am(III) was carried out using a capillary electrophoretic technique in a mixed solvent (CH₃OH/H₂O) system containing thiocyanate ion. First, the formation constants (β_n) between thiocyanate ion and Eu(III) or Am(III) were investigated in mixed solvent solutions of different ionic strength by a back-extraction technique using bis (2-ethylhexyl) hydrogen phosphate-toluene. The values of β_n for Am(III) were considerably larger than that of Eu(III). Next, it was examined for the moving velocities (v) of Eu(III) and Am(III) species in 1.10 M (H⁺, Na⁺)(SCN⁻, ClO₄⁻) mixed solvent solution by a paper electrophoretic technique. The values of v of Eu(III) were considerably larger than those of Am(III) in the mixed solvent solution contains 0.20 ~ 0.60 M [SCN⁻]. On the basis of the data of β_n and v , the capillary electrophoresis separation of Am(III) from Eu(III) was tried in 0.20 M NaSCN mixed solvent solution ($X_{\text{MeOH}} = 0.40$) system. The results showed that Am(III) was separated almost completely from Eu(III).

3P23 : Study on f-block element ions by solvent extraction using cyclic polythiaether

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It is expected that 1,4,7,10,13,16-hexathiacyclooctadecan ([18]aneS6) might have a more selective complexation toward actinides(III) than lanthanides(III), based on a slight difference of the softness between actinides(III) and lanthanides(III). Then, the effect of [18]aneS6 was examined by ion-pair extraction of actinides(III) and lanthanides(III)

using tetrakis[3,5-bis(trifluoro-methyl)phenyl]borate(TFPB⁻) of a hydrophobic anion. Actually, the extraction behaviors of ^{152,154}Eu(III) and ²⁴¹Am(III) in the region pH=3.1~6.3 were investigated using [18]aneS6 and TFPB⁻ dissolved into 1,2-dichloroethane. The distribution ratios of both metal ions in trace concentrations showed the highest values in the vicinity of pH=3.3, respectively. It was shown that the distribution ratios of Eu(III) and Am(III) were dependent on the concentration of [18]aneS6. The effect of other counter anions except TFPB⁻ has also been examined. Consequently, it was found that the ions of Eu(III) and Am(III) can be more effectively extracted by using TFPB⁻ than the other counter anions.

3P24 : Adsorption behavior of f-elements on tertiary pyridine and quaternary pyridinium resins

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The chromatographic separation experiments of lanthanides and actinides were carried out by using the tertiary pyridine resin and the quaternary pyridinium resin with the nitric acid / methanol mixed solution and the hydrochloric acid / methanol solution. The tertiary pyridine resin has two functions, one is a function of weakly basic anion exchanger, and another is a function of soft donor ligand. Meanwhile, the quaternary pyridinium resin has only one function of strongly basic anion exchanger. From a comparison of adsorption behaviors of f-elements on these two types of resin, it was concluded that the f-elements adsorb on the resin by mechanism of ion exchange in nitric acid system and on the other hand in hydrochloric acid system, the adsorption mechanism is attributable to coordinate bonds of pyridine and f-elements.

3P25 : Determination of transfer energy of actinide ions between aqueous and organic solution phases

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The Gibbs energy for transfer (ΔG_{tr}°) of an ion at the interface between aqueous and organic solution phases is a fundamental parameter for distribution of the ion on solvent extraction. The purpose of present work is to determine ΔG_{tr}° of actinide ions of various oxidation state. At first, the ΔG_{tr}° of H⁺ ion was measured by voltammetry for the ion transfer at the micro-interface between immiscible electrolyte solutions. The ΔG_{tr}° of actinide ions was determined based on comparison of extractability of H⁺ and actinide ions after distribution equilibrium. Nitrobenzene, 1,2-dichloroethane, chloroform, acetophenone, benzonitrile and *o*-nitrophenyl octyl ether have been employed as organic phase. Relation between ΔG_{tr}° determined and solvent property is discussed.

3P26 : Solvent extraction of actinide ions by N,N-dioctyldiglycolamic acid

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N,N-Dioctyldiglycolamic acid (DODGAA) was synthesized from diglycolic anhydride and N,N-dioctylamine. Solvent extraction of Am(III), Pu(IV), Np(V), and U(VI) from aqueous HNO₃ solution was performed using 0.1 M DODGAA dissolved in n-dodecane including 10 vol% 1-octanol. It was found that DODGAA had high extractabilities for Am(III), Pu(IV), and U(VI) from dilute HNO₃ into n-dodecane. The distribution ratios of these actinide ions decrease with increasing HNO₃ concentration, indicating that DODGAA acts as an acidic type ligand; that is, the metal cations are complexed with the carboxylate anions, which are formed by deprotonation of the carboxyl group of DODGAA at low acidity.

3P27 : Separation of Am(III) by Nitrogen-Donor Ligand

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Soft-donor extractants containing sulfur- or nitrogen-donor are applicable to the separation of trivalent actinides (An(III)) from lanthanides (Ln(III)) by solvent extraction method. We designed two ligand systems which are expected to efficiently separate An(III) from Ln(III). The one is "Oligo-pyridine" ligand containing multiple pyridine moieties. The other is "Chirality-controlled" ligand containing chiral centers in the ligand. We synthesized more than thirty ligands and found out the ability on the separation of An(III) from Ln(III). In this presentation, we will discuss about the comprehensive results on the separation ability of An(III) from Ln(III) by means of "Oligo-pyridine" and "Chirality-controlled" ligands, and the extraction equilibrium of An(III) with selected tripodal ligands in detail.

3P28 : Synthesis and crystal structures of various Uranyl(VI) complexes coordinated by acac

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

Single crystals of [UO₂(acac)₂(imH)], [UO₂(acac)₂(pyridine-4-aldxime)] and (2,2'-bpy)₂H[UO₂(acac)(NO₃)₂] have been synthesized, and the three crystal structures have been determined by using X-ray CCD. The [UO₂(acac)₂(imH)] and [UO₂(acac)₂(pyridine-4-aldxime)] complexes exhibit pentagonal-bipyramidal geometry about the uranyl(VI) ion. The both complexes involve two bidentate acac ligands and one monodentate N-heterocyclic ligand. The (2,2'-bpy)₂H[UO₂(acac)(NO₃)₂] is hexagonal-bipyramidal geometry about the uranyl(VI) ion. The complex involves one bidentate acac and two bidentate nitrate ions. The 2,2'-bpy molecule is not coordinated by uranyl(VI) ion and is protonated to (2,2'-bpy)₂H⁺ ion.

3P29 : The interaction between trivalent f-block element ions and iodide ion

Hayashi, K.¹, Mori, T.¹, Yanaga, M.¹, Satoh, I.², Suganuma, H.¹ (¹Radiochem Res. Lab., Fac. Sci., Shizuoka Univ., ²IMR, Tohoku Univ.)

The formation constants of solvent-shared ion-pair of lanthanides(III)-I⁻ and Am(III)-I⁻ in a mixed solvent of methanol and water was studied by a back-extraction technique. The organic phase in the extraction procedure is a

toluene solvent containing bis(2-ethylhexyl)hydrogen-phosphate with a trace amount of radionuclide of lanthanides(III) or Am(III) and the aqueous phase of the mixed solvent contains 1.0 mol dm⁻³ (H, Na)(I, ClO₄). In a methanol mole fraction (X_S) < 0.31, the formation constants of Eu³⁺-I⁻ (β_1 (Eu)) are a somewhat larger than β_1 (Am), but the values of β_1 (Eu) at $X_S = 0.31$ and 0.40 are approximately equal to the values of β_1 (Am). However, Am(III) at $X_S = 0.31$ and 0.40 possesses the values of β_2 (Am), the other hand, the β_2 (Eu) were not measured. Furthermore, it was found that the value of β_1 of lanthanides(III) increases with an increase of atomic number.

3P30 : Preparations and crystal structures of 8 coordinate uranyl(VI) complexes having macrocyclic ligands derived from pyrroledicarboxialdehydes and diamines

Komagine, J., Takeda, M., Takahashi, M. (Dept. Chem., Toho Univ)

Six 8-coordinate uranyl(VI) complexes with macrocyclic Schiff base ligands derived from 2,6-pyrroledicarboxialdehyde and diamines are prepared and the crystal structures for two of them are determined focusing on the relation between the size of the ligands and U-N bond distances. No difference in average uranyl bond distances and bond angles are observed between [UO₂(bipytn)](a) and [UO₂(bipydmtn)](b). U-N bonds of these complexes are, however, not equal; the U-N(pyrrole) bonds [2.45(a), 2.44(b) Å] are much shorter than the U-N(imine) bonds [2.67(a), 2.67(b) Å].

3P31 : Positive muons in β -tricalcium phosphate

Kakuyama, T.,¹ Kubo, M. K.,¹ Tsuruoka, Y.,¹ Koda, A.,² Nishiyama, K.² (¹International Christian Univ., ²KEK)

Stable hydrogen atoms were observed at room temperature by an ESR study in X-ray irradiated tricalcium phosphate (β -Ca₃(PO₄)₂, TCP). The hydrogen was estimated to occupy only one site based on the interpretation of the superhyperfine splitting of the ESR spectrum. The formation and sites of muonium, a light isotope of hydrogen, and the behavior of diamagnetic muons in polycrystalline TCP were investigated by the μ SR method. In 0.59 mT transverse field at 294 K, a spin precession at 8.0 ± 1.3 MHz was observed and identified as the precession of the muon spin in muonium. The relaxation rate of the muon spin polarization was 3.8 ± 1.1 μ s⁻¹ and the yield was 6.8 ± 4.1 %. Muonium was also observed at 4.6 K and 39.9 K. The relaxation rate of the muon spin polarization in diamagnetic muon increased at low temperatures. In 6.0 mT at 294 K, the muon spin polarization at 4.8 ± 1.0 μ s⁻¹ was observed and estimated as another diamagnetic muon species.

3P32 : basic study for the gas-phase chemical reaction of muonic atoms

Nakatsuka, T.,¹ Ninomiya, K.,¹ Sugiura, H.,¹ Sato, W.,¹ Yoshimura, T.,¹ Kubo, M. K.,² Matsumura, H.,³ Miura, T.,³ Nishiyama, K.,³ Shinohara, A.¹ (¹Osaka Univ., ²International Christian Univ., ³KEK)

A captured muon exists for considerably long time on an 1s orbital. Hence, muonic atoms possibly behave as chemical species of Z-1 atom. We have investigated the electronic structure of the muonic atoms. In addition, a preliminary experiment to research chemical reactivity of muonic atoms was also performed in this research. The measurements were performed at the Muon Science Laboratory in High Energy Accelerator Research Organization. In order to investigate the electronic structure of the muonic atoms, targets of the elements ranging from Sn to U were irradiated with muons. Additionally, as a preliminary experiment to research chemical reactivity of muonic atoms, we irradiated Ar gas, CH₄ gas and a mixture of the Ar and CH₄ gases with muon beam and attempted to detect chemical luminescence emitted by the gas-phase chemical reaction of muonic atoms.

3P33 : Mössbauer spectra of tris(2-pyridylmethyl)-amine-iron complexes

Sakai, H., Yamawaki, S., Koura, N., Miyake, S., Fujii, S. (Dep. Chem., Konan Univ.)

A series of iron(II) complexes with the tetradentate tris(2-pyridylmethyl)amine (TPA) ligand have been prepared and characterized. Variable-temperature Mössbauer measurements revealed that Fe(II)(TPA)(NCS)₂ exhibited a spin-crossover phenomenon of the spin equilibrium type. The color of the complex changes from yellow in the high spin state to dark red in the low spin state. The complex [Fe(II)(TPA)(bpy)](BPh₄)₂ with the strong bipyridine ligand exhibited a low spin state at 80K, whereas [Fe(II)(TPA)(H₂O)₂](BPh₄)₂ and Fe(II)(TPA)Cl₂ with weak ligands showed high spin states. It is established from the Mössbauer parameters and a calculation of the optimum structure that the [Fe(II)(TPA)Cl](BPh₄) complex has a trigonal bipyramidal iron center.

3P34 : Spin crossover behaviors of Fe(II) complexes of polydentate ligands containing imidazole group

Iijima, S.,¹ Niwa, O.,¹ Hagiwara,² H., Torigoe, H.,² Matsumoto, N.² (¹AIST, ²Kumamoto Univ.)

Spin crossover behaviors of iron(II) complexes of several polydentate ligands containing imidazole group were investigated by ⁵⁷Fe Mössbauer spectroscopy. The Schiff base HL prepared from 2-(2-aminoethyl)pyridine and 2-methyl-4-formylimidazole afforded two types of Fe(II) complexes [Fe(HL)₂](PF₆)₂·0.5H₂O as a tridentate ligand and [Fe(HL)₃](PF₆)₂ as a bidentate ligand. While [Fe(HL)₂](PF₆)₂·0.5H₂O exhibited a gradual low spin-high spin transition between 4.2 K and 200 K, the spin transition in [Fe(HL)₃](PF₆)₂ occurred abruptly around 170 K. The 1D-chain structure of the latter complex consisting of NH...N hydrogen bonds between the imidazole and pyridine moieties would contribute to the rapid change of the spin state.

3P35 : Photoinduced spin transition phenomena for Fe(III) spin-crossover complex

Hiki, K., Hayami, S., Maeda, Y. (Kyushu Univ.)

Spin crossover phenomenon occurs in some six coordinate

first-row transition metal complexes as the result of an electronic instability driven by external constraints. For iron spin crossover complexes, ⁵⁷Fe Mössbauer spectroscopic method is very useful to observe electron configuration, chemical bonding and so on of center iron ions. Some iron(II) complexes show light induced excited spin state trapping (LIESST) effect. LIESST effect is occurred by photoexcited complex is trapped in metastable HS phase. In 10 years ago, it is thought that only some iron(II) spin crossover complexes show LIESST effect and other complexes are impossible to do. However, Hayami and co-workers observed LIESST effect in iron(III) spin crossover complex in 2000. Fig. shows magnetic susceptibility and time dependence metastable HS-state to LS-state relaxation curve for [Fe(pap)₂](ClO₄). This complex exhibits high T_{LIESST} (= 90K) and long life time of metastable HS phase, the tunneling rate constant $k_{HL}(T \rightarrow 0) = 2.8 \times 10^{-8} \text{ s}^{-1}$. In this presentation, from relaxation experiment and comparing with Fe(II) LIESST complexes or Fe(III) spin-crossover complexes, we approach to the reason of why [Fe(pap)₂](ClO₄) exhibits LIESST effect.

3P36 : Photo-switching of ferromagnetic FePt nanoparticles

Einaga, Y.,¹ Suda, M.,¹ Iyoda, T.² (¹Keio Univ., ²Tokyo Inst. Tech.)

We have reported several examples of novel photo-switching magnetic systems containing photochromic compounds and magnetic materials. That is, the magnetic properties can be controlled by photoisomerization of the photochromic compounds. In order to realize reversible photo-switching of magnetization at room temperature, we have focused on a surface modification of nano-scale magnetic particles and a combination of photochromic azobenzene-containing amphiphilic compounds and the magnetic nanoparticles as magnetic materials that can function at room temperature. Here we have designed reversible photo-switchable ferromagnetic FePt nanoparticles whose surfaces were coated with azobenzene derivatized ligands. These composite nanoparticles showed ferromagnetic behavior even at room temperature. The photoisomerization brought changes in the electrostatic field around the core-FePt nanoparticles. As a result, we have succeeded in controlling the magnetic properties of these ferromagnetic composite nanoparticles by alternating photoillumination in the solid state at room temperature.

3P37 : Characterization of iron-nanoparticles in carbon nanotubes by Mössbauer spectroscopy

Nagata, K.,¹ Tajima, C.,² Muramatsu, H.,² Kim, Y. A.,³ Endo, M.³ (¹Grad. School of Edu., Shinshu Univ., ²Fac. of Edu., Shinshu Univ., ³Fac. of Eng., Shinshu Univ.)

Among the various types of synthesis methods for carbon nanotubes (or nanofibers), a catalytic thermal chemical vapor deposition (CVD) synthesis method has been considered as a promising method for large-scale production of carbon nanotubes, especially using the floating reactant method established by Prof. M. Endo et al. When composing carbon nanotubes (CNTs), iron-catalyst nanoparticles (which catalyze the formation of CNTs) get mixed as a by-product in the final product. In order to detect and characterize iron-nanoparticles, the Mössbauer spectra have been measured for five different CNTs samples; A: Cup-stacked

Nanotubes (as-grown), B: Multi-wall Nanotubes (as-grown), C: Multi-wall Nanotubes (as-grown), D: Multi-wall Nanotubes (high temp. treatment = 2800°C), E: Cup-stacked Nanotubes (high temp. treatment = 2800°C). The results show that the iron existed as Fe, iron oxides, iron sulfides, and iron carbides, showing magnetically splitting peaks, only for CNT-A sample. For other samples, however, no magnetic component exists in their spectra, but quadrupole doublets in the vicinity of 0 mm/s still exist. This fact suggests that most of by-products can be removed by purification or high temperature treatment, while iron-nanoparticles included inside the CNTs cannot.

3P38 : Effects of droplets on spin-orientation of iron films produced by laser-deposition

Yamada, Y., Namiki, K., Kato, H. (Tokyo Univ. of Sci.)

Laser-deposition is very useful technique to produce thin films of various materials. Laser-evaporation produces not only atoms but also small particles depending on the condition; the particles are called as droplets. Deposition process is complicated because laser-evaporated materials have high energy and have possibility to react with substrate materials. Mössbauer spectroscopy is very useful to investigate magnetic properties or spin orientation of the iron based materials. Here, the effects of droplets on the spin-orientation are studied. Laser light from a YAG-laser (532 nm, 100 mJ/pulse, 5 ns) was focused by a convex lens onto a block of enriched ^{57}Fe metal in a vacuum vessel. Mössbauer spectra of the films on the substrates were measured at room temperature in transmission geometry. We also examined the films using scanning electron microscopy. Laser-deposition of Fe on Al substrate produces α -Fe films with spin oriented to parallel to the surface, while part of the Fe atoms migrates into the substrate. The laser-deposited Fe film having a lot of droplet prevents the migration into the substrate though the spin orientation is random.

3P39 : A neutron in-beam Mössbauer spectrum of iron disulfide at low temperature

Tsuruoka, Y.¹, Kubo, M. K.¹, Kobayashi, Y.², Yamada, Y.³, Takayama, T.⁴, Watanabe, Y.^{4,5}, Sakai, Y.⁴, Shoji, H.⁶, Sato, W.⁷, Shinohara, A.⁷, Matsue, H.⁸ (¹ICU, ²RIKEN, ³Tokyo Univ. Sci., ⁴Daido Inst. of Tech., ⁵Saint-Gobain, ⁶TMU, ⁷Osaka Univ., ⁸JAEA)

Neutron in-beam Mössbauer spectroscopy is a useful tool for characterization of the products after neutron capture reactions nondestructively. The Mössbauer γ -ray from ^{57}Fe arising from the $^{56}\text{Fe} (n, \gamma) ^{57}\text{Fe}$ reaction in iron disulfide (FeS_2) was measured. Powder of FeS_2 (pyrite) was pressed to form a thin disk of 31 mm in diameter and about 150 mg cm^{-2} in thickness. The disk was oriented both to the neutron beam and the detector at an angle of 45 degrees and measured at 78K in a liquid nitrogen cryostat. We will present a spectrum of pyrite at 78K with a good signal to noise ratio.

3P40 : Mössbauer resonance absorption on FePSe_3 and FePTe_3

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We have already reported the first steps towards a proof-of-principle experiment, demonstrating electro-magnetically induced transparency (EIT) with gamma radiation for the Mössbauer effect in the mineral siderite FeCO_3 using the level-crossing technique. For the single crystal of FeCO_3 synthesized in a laboratory, however, it seems so hard to make large single crystal enough to be used as an absorber. Then, we tried to seek another absorber possible to use in the same type of experiment. Although the first candidate we had tried was the single crystal of FePS_3 , the internal-magnetic-field strength was not enough to make the absorption lines of $(-3/2 \leftrightarrow -1/2)$ and $(1/2 \leftrightarrow -1/2)$ merge into one single line, in order to observe EIT. Then, for the next step, we have tried to synthesize the single crystal of FePSe_3 or FePTe_3 . From a series of Mössbauer measurements using FePSe_3 and FePTe_3 absorber, it seems that both materials cannot be used as an absorber in a proof-of-principle experiment demonstrating EIT. Since the experiments for FePSe_3 and FePTe_3 are in progress and both materials are not pure or not identified yet, nothing is conclusive.

3P41 : Mössbauer study on IZO doped with diluted ^{57}Fe

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Powder of IZO (In:Zn=1:1) doped with 1% ^{57}Fe was first prepared by a sol-gel method and characterized by VSM, XRD and Mössbauer spectrometry. It was confirmed from SEM-EDX analysis that the powder obtained is uniform in distribution of elements contained. XRD pattern with broad peaks showed that the powder is like amorphous. VSM showed weak ferromagnetic susceptibility. In Mössbauer spectrum, doublet and magnetic relaxation peaks were observed. These results are discussed.

3P42 : On the formation of akaganeite in the presence of different cations

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Akaganeite, $\beta\text{-FeOOH}$, samples were prepared by hydrolysis of FeCl_3 solutions in the presence of Al^{3+} , Cr^{3+} , Cu^{2+} and Ti^{4+} ions at different concentrations. XRD, FTIR and Mössbauer spectrometry show that akaganeite is the only phase being formed, i.e. they do not promote the formation of another phase. Very small differences in some of the crystallographic and hyperfine parameters for samples obtained in the presence of Al^{3+} , Cr^{3+} , Cu^{2+} ions at concentrations lower than 10 mol% were observed. However at 30 mol%, the presence of Al is able to reduce the average grain size and the magnitude of some hyperfine parameters of the final products. One does conclude that the present chemical route allows only rather small amounts of Al, Cr and Cu to enter in the structure of akaganeite. On the other hand, Ti^{4+} drastically affects the physical properties of the akaganeite: its presence decreases the grain sizes, reduces all the unit cell parameters and introduces defects into the crystallographic structure. The types of defects presented in the samples require further investigation.

3P43 : Iodine-127 Mössbauer spectra of (dihaloiodo)benzenes

Takahashi, M., Higashidate, M., Takeda, M. (Dep. of Chem., Toho Univ.)

Iodine-127 Mössbauer spectrum of a hypervalent iodine(III) compound IolIF_2 (Tol = $p\text{-CH}_3\text{-C}_6\text{H}_5$) is measured at 20 K and its electronic state is compared to that of IolICl_2 . The estimated valence electron population of iodine(III) atom ($5s^{1.87} 5p_x^{0.38} 5p_y^{1.13} 5p_z^{2.0}$) is in good agreement with that of calculated by *ab initio* calculation, indicating that the fluorine atom attracts much more electron (about 0.88 e) from iodine atom (cf 0.64 e in IolICl_2). A detail analysis of the ^{127}I Mössbauer parameters suggests that 5s electrons are also participate in the hypervalent bond and the fluorine atoms also interact with the benzene ring. The x-ray crystal structure of IolIF_2 at 218 K are also described.

3P44 : ^{155}Gd Mössbauer spectroscopic investigation of 3d-4f and 4f-4f' dinuclear complexes containing Gd (III)

Ayabe, T.,¹ Takahashi, M.,¹ Takeda, M.,¹ Costes, J. P.,² (¹Toho Univ., ²CNRS)

^{155}Gd Mössbauer spectra for dinuclear complexes of Gd^{3+} and lanthanide (III) ion having a Schiff base ligand (L) derived from tren and 3-ethoxysalicylaldehyde were measured at 12K. Mössbauer spectra of these complexes showed quadrupole split spectra. Two kinds of isomer shift values are observed; i.e., $\delta = 0.57 - 0.60 \text{ mm s}^{-1}$ for LGdLn' ($\text{Ln}' = \text{Ce}, \text{Nd}, \text{Eu}, \text{Gd}$) and $\delta = 0.64 - 0.66 \text{ mm s}^{-1}$ for LLnGd ($\text{Ln} = \text{Dy}, \text{Er}, \text{Yb}$). The former values are very close to those of nitrogen-containing Gd (III) complexes ($\delta = 0.59 - 0.62 \text{ mm s}^{-1}$) such as edta and cyano-bridged complexes. The latter values are on the other hand, in the range of oxygen-coordinating Gd (III) complexes ($\delta = 0.64 - 0.65 \text{ mm s}^{-1}$) such as polyethylene glycol Gd (III) complexes. ^{155}Gd Mössbauer spectra for LLnLn' complexes indicate that the smaller lanthanide ion Ln is in the N_4O_3 site and the larger ion Ln' in the O_{10} site.

3P45 : Preparation and Mössbauer spectroscopic study of 4-cyanopyridine coordinated neptunyl complex

Nakada, M.,¹ Komagine, J.,² Takahashi, M.,² Kitazawa, T.,² Nakamura, A.,¹ Takeda, M.,² (¹JAEA, ²Toho Univ.)

We have studied chemical and physical properties of Np compound using ^{237}Np Mössbauer spectroscopy. Recently, pyridine and bipyridine coordinated neptunyl complexes were synthesized and measured by ^{237}Np Mössbauer spectroscopy. In the present work, 4-cyanopyridine coordinated neptunyl complex was synthesized and measured by ^{237}Np Mössbauer spectroscopy. 4-Cyanopyridine coordinated uranyl complex was synthesized for confirming a synthetic method. This complex was measured by X-ray diffraction. A coordination number of U atom was 7. 4-Cyanopyridine coordinated neptunyl complex was synthesized according to the uranyl complex synthetic method. ^{237}Np Mössbauer spectrum of the neptunyl complex was measured at low temperature. We observed the spectrum with 16 absorption lines at 10 K.

3P46 : Mössbauer spectra of U-Eu mixed oxide

Masaki, N., Nakada, M., Otake, H. (JAEA)

We have studied chemical properties of fluorite-type structure compounds by using powder XRD and Mössbauer spectroscopy. ^{151}Eu Mössbauer spectroscopic study on mixed oxide, $\text{U}_{1-y}\text{Eu}_y\text{O}_{2-x}$ revealed that oxygen coordination number around Eu^{3+} was 8 for $0 < y < 0.5$ and decreased smoothly with increase in y for $y > 0.5$. ^{238}U Mössbauer spectroscopy was applied to this system. The γ -ray source used for ^{238}U Mössbauer spectroscopy was $^{242}\text{PuO}_2$ (^{242}Pu : 99.99%). 44.9 keV ^{238}U Mössbauer γ -ray is distinguished from the 59.5 keV γ -ray from the decay of ^{241}Am isotope which is the daughter of isotopic impurity ^{241}Pu by using a Ge detector. The magnetic splitting of ^{238}U Mössbauer spectra for UO_2 was observed as obvious broadening of the line shapes below the Neel temperature, 30K.

4P01 : (withdrawal)

4P02 : Development of gridded ionization chamber for measuring atomic number of fission fragments

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

In order to investigate the mechanism of asymmetric mass division in low energy fission of actinides, the detector for measuring an atomic number (Z) for fission fragments has been developed. Because the atomic number is closely related to energy losses of fragments, the gridded ionization chamber with divided anode is useful for this purpose. The detector was designed and optical conditions such as the distance and electric potential between electrodes were searched using alpha particles from ^{252}Cf . The total energy and energy losses of fission fragments from ^{252}Cf were measured under the obtained conditions. It was found that fission fragments lost most of the kinetic energy in the beginning of their range. This behavior agrees qualitatively with results of simulation by TRIM code. In the presentation, the results of energy measurements under various conditions will be shown and discussed.

4P03 : Study on the synthesis of heavy elements using the $\text{Ba} + ^{16}\text{O}$ and $\text{La} + ^{16}\text{O}$ reaction systems

Arai, M.,¹ Kinoshita, N.,¹ Nanri, T.,² Yokoyama, A.,¹ Takahashi, N.,³ (¹Grad. School Nat. Sci. Tech., Kanazawa Univ., ²Fac. Sci., Kanazawa Univ., ³Grad. School Sci., Osaka Univ.)

In order to study the nuclear fusion of deformed nuclei with heavy ions, we adopted lanthanoid targets, which are known to be deformed as actinoides. We performed bombardment of Ba and La targets with ^{16}O ions at the Research Center for Nuclear Physics, Osaka University, and measurement with a Ge detector, a silicon semiconductor detector, and a photon electron rejecting alpha liquid scintillation counter to obtain cross sections of the products. They were compared with a theoretical excitation function. As a result, it was found that the threshold energy of fusion reaction was higher than the value expected by theory.

4P04 : Measurement of ultraviolet, visible and infrared light originating from de-excitation of $^{229\text{m}}\text{Th}$

Nakashima, K.,¹ Kasamatsu, Y.,^{1,2} Sato, W.,¹ Kikunaga, H.,^{3,6} Takamiya, K.,⁴ Mitsugashira, T.,⁵ Nakanishi, T.,⁶ Ohtsuki, T.,⁷ Shinohara, A.¹ (¹Osaka Univ., ²JAEA, ³RIKEN, ⁴Kyoto Univ., ⁵Inst. Materials Res., Tohoku Univ., ⁶Kanazawa Univ., ⁷Grad. School Sci., Tohoku Univ.)

An isomer of ^{229}Th , $^{229\text{m}}\text{Th}$, is expected to show an interesting decay property because of its extremely low-lying excitation energy. If $^{229\text{m}}\text{Th}$ mainly disintegrates through an electronic bridge process, the decay rate of $^{229\text{m}}\text{Th}$ would change depending on the chemical state of $^{229\text{m}}\text{Th}$. In our previous work, photons emitted from $^{229\text{m}}\text{Th}$ in HCl solution and in HNO_3 solution were not detected. Expecting observation of photons by changing chemical condition, we tried to detect optical photons for $^{229\text{m}}\text{Th}$ hydroxide precipitate samples and $^{229\text{m}}\text{Th}$ fluoride precipitate samples in this work. Although photon emission was observed, we found that detected photons were originated from radiation from matter other than $^{229\text{m}}\text{Th}$. In the expectation that the photon energy from $^{229\text{m}}\text{Th}$ is lower than reported values, a photon measurement apparatus was developed employing a photomultiplier that has high sensitivity for photons of a red-infrared region, and we tried to observe photon emission with the apparatus for $^{229\text{m}}\text{Th}$ sample.

4P05 : Systematical properties of spontaneous fission for superheavy nuclei and a limit of existence of nuclei

Koura, H. and Chiba, S. (JAEA)

We present a chart of nuclear decay modes for α decay, β decay, proton emission, and spontaneous fission ranging from light nuclei to superheavy nuclei between neutron- and proton-drip lines with the use of the KTUY nuclear mass formula to estimate decay rates of the above ones. The standard deviation of this mass formula from known masses is 0.67 MeV, and below 0.4 MeV from some separation energies. The WKB methods are applied to calculate decay rates without β -decay one, which is calculated with the gross theory. With these calculations, we will show some theoretical results on decay properties in the superheavy nuclidic region. We also calculate total half-lives for nuclei in the "island of stability for the superheavy nuclei" and obtain an α -decay-dominant nucleus with the longest half-life on the β -stability line of our mass formula in the order of 100 years with a certain ambiguity. Furthermore, we also estimate the nuclei beyond the superheavy ones and will discuss the next "island of stability" and the limit of existence of nuclei which is given not by proton emission, but by fission.

4P06 : Development of ion source for JAEA-ISOL-I

Osa, A., K. Sato, T., Ichikawa, S. (JAEA)

According to the JAEA-KEK joint radioactive nuclear beam (RNB) project, we have improved a surface-ionization type integrated-Uranium-target-ion source (U-SIS) system to separate neutron-rich indium nuclei with particle-induced fission of ^{238}U . Yields and release times of mass-separated

indium nuclei were examined. An overall efficiency for ^{126}In separation was 0.08% by a previous version of U-SIS. We considered this very low efficiency was caused by the low temperature of a thick ionizer. Therefore, we used a thin ionizer for the improved version of U-SIS. A uranium carbide target was prepared at a uranium density of $630\text{-mg/cm}^2\text{U}$. The target was bombarded with a 33-MeV proton beam with intensity of about 100 nA. For the separation yield measurements of fission products, the mass separated products were collected on an aluminized Mylar tape in a tape transport system and were periodically transported to a measuring position where an HPGe detector was placed for γ -ray measurements. Separation yields of indium nuclei were derived from γ -ray measurements. The overall efficiency for ^{126}In separation was about ten times improved. Release time of In ion was also measured.

4P07 : Development of ion source for JAEA-ISOL-II

Sato, T. K., Osa, A., Tsukada, K., Asai, M., Ichikawa, S. (JAEA)

Isotope separation on-line (ISOL) with unambiguous mass identification and rapid separation is a powerful method for study of short-lived trans-plutonium isotopes. So far, we installed a gas-jet coupled thermal ion source in the JAEA-ISOL, and neutron-deficient trans-plutonium isotopes included the new isotopes ^{233}Am , ^{237}Cm and ^{241}Bk , were successfully mass-separated and these decay properties were studied. To extent this research activities in the region of other trans-plutonium isotopes, we have been studied a thick target ion-source technique for increment separation yield of an isotope of interest. Recently, we successfully mass-separated ^{239}Am isotope produced in the heavy-ion fusion evaporation reaction with the intensity of 1.3×10^3 atoms/s from thick uranium target. The observed beam intensity of ^{239}Am isotope is about 10 times higher than that expected with the gas-jet coupled thermal ion source system.

4P08 : Studies on the heavy element productions at RIKEN for chemical characterization of seaborgium

Takabe, T.,¹ Saika, D.,¹ Matsuo, K.,¹ Tashiro, Y.,¹ Ooe, K.,¹ Kuribayashi, T.,¹ Yoshimura, T.,¹ Toyoshima, A.,² Kikunaga, H.,³ Kaji, D.,³ Haba, H.,³ Kudo, H.,⁴ Mitsugashira, T.,⁵ Shinohara, A.¹ (¹Osaka Univ., ²JAEA, ³RIKEN, ⁴Niigata Univ., ⁵Tohoku Univ.)

The gas-jet coupled heavy element production system was installed on the beam line of the RIKEN K70 AVF Cyclotron for investigation of chemical properties of Seaborgium (Sg, $Z=106$). Prior to the production of the nuclide ^{265}Sg in the $^{248}\text{Cm}(^{22}\text{Ne},5n)$ reaction, the production cross sections of the heavy nuclides ^{255}No and ^{261}Rf produced by the $^{238}\text{U}(^{22}\text{Ne},5n)$ and $^{248}\text{Cm}(^{18}\text{O},5n)$ reactions, respectively, were measured. The irradiation test with the high-intensity ^{22}Ne beams on the ^{238}U target and the evaluation of background α -particles in the $^{248}\text{Cm}(^{18}\text{O},5n)^{261}\text{Rf}$ experiment were also performed.

4P09 : Startup of on-line chemistry at Research Center for Nuclear Physics, Osaka University

Tashiro, Y.,¹ Saika, D.,¹ Kitamoto, Y.,¹ Matsuo, K.,¹ Takabe,

T.,¹ Kuribayashi, T.,¹ Ooe, K.,¹ Yoshimura, T.,¹ Sato, W.,¹ Takahashi, N.,¹ Takahisa, K.,² Shinohara, A.¹ (¹Osaka Univ., ²RCNP)

Recently, an irradiation system was newly installed at the K course of the AVF cyclotron facility of Research Center for Nuclear Physics (RCNP), Osaka University. The irradiation chamber is connected with a gas-jet transport system for rapid experiments. We also fabricated a rapid solvent extraction apparatus using a multi-track microchip connected with the gas-jet transport system for the purpose of studying extraction behavior of heavy actinide elements. Test experiments using lanthanide radioisotopes were carried out. At first, the transport efficiency of the lanthanide isotopes were studied in various conditions. The transport efficiency of the gas-jet system was optimized at the temperature of KCl aerosol clusters of 640 °C and at the He flow rate of 2.5 L/min. The transport efficiency was varied between 10% and 60%. We currently started studying the solvent extraction behavior of Cf and Fm with this system.

4P10 : Rapid solvent extraction experiment of No²⁺ with multi-track microchips

Tashiro, Y.,¹ Saika, D.,¹ Kitamoto, Y.,¹ Matsuo, K.,¹ Takabe, T.,¹ Kuribayashi, T.,¹ Ooe, K.,¹ Yoshimura, T.,¹ Sato, W.,¹ Takahashi, N.,¹ Toyoshima, A.,² Haba, H.,³ Shinohara, A.¹ (¹Osaka Univ., ²JAEA, ³RIKEN)

We have developed an online rapid chemical operation system for heavy actinide elements. On this study, multi-track microchips were adopted for the rapid extraction experiment. We performed the online experiment using AVF cyclotron at RIKEN. A ²³⁸U target was irradiated with a 112.4 MeV/Nucl. ²²Ne beam, and ²⁵⁵No nuclei were produced by the ²³⁸U(²²Ne,5n)²⁵⁵No reaction. The products recoiling out of the target were transported to the laboratory for chemical experiments, dissolved in aqueous solution, and then fed into the multi-track microchips. The ²⁵⁵No isotope was rapidly extracted in the multi-track microchip by means of 0.01 M HDEHP(Di(2-ethylhexyl)phosphoric acid)-toluene solution as the extractant. And α particles emitted from the isotope detected 40 counts in total. The distribution ratio of the rapid extraction of ²⁵⁵No using multi-track microchip showed the dependence of the ionic radius in the order $\text{Ca}^{2+} < \text{No}^{2+} < \text{Sr}^{2+}$.

4P11 : Basic study on gas phase chemistry using carbon cluster transport system

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

During the past several years, our group has studied the volatile properties of the compounds of the group-4 elements including Rf, especially tetrachloride compounds, using an isothermal gas chromatographic method. When He/KCl and HCl were used as the transportation and the reaction gas respectively, serious problem arose because of the generation of the nonvolatile chloride oxides by the residual oxygen in the system. In this work, the carbon cluster transport system was newly applied in order to eliminate the residual oxygen in the system. Using the fission fragments from ²⁵²Cf, it was found that the transport efficiency was about 50%, and the volatile Zr compound was also measured. From this result, it is suggested that this system is applicable to the gas phase chemistry experiments. The result of the chromatography of Zr compound will be discussed in the presentation.

4P12 : Development of an electrochemistry apparatus for atom-at-a-time chemistry

Toyoshima, A.,¹ Kasamatsu, Y.,¹ Tsukada, K.,¹ Haba, H.,² Shinohara, A.,³ Nagame, Y.¹ (¹JAEA, ²RIKEN, ³Osaka Univ.)

An electrochemistry apparatus for an atom-at-a-time chemistry was developed. A surface of glassy carbon fibers filled in a vycor-glass tube to be used as a working electrode was chemically modified by Nafion perfluorinated ion exchange resin so that elution behavior of radiotracers from the electrode is according to their charge states and ionic radii. Some of oxidizable elements such as cerium (Ce) are oxidized on the working electrode according to their own oxidation potentials, and the resulting oxidized species such as Ce⁴⁺ are separated from not oxidized ones (Ce³⁺). Oxidation/separation behavior of the ¹³⁹Ce radiotracer of 10¹⁰ atoms in 0.1 M α -hydroxyisobutyric solution was studied using the apparatus. Elution positions of ¹³⁹Ce shifted from that of Ce³⁺ to that of Ce⁴⁺ at around 0.75 V (vs. Ag-AgCl reference), and the electric potential of 0.75 V was almost equal to that determined in cyclic voltammetry of 0.001 M Ce. Therefore, it was concluded that ¹³⁹Ce in the trivalent state is successfully oxidized to the tetravalent one using the present apparatus on a tracer scale.

4P13 : TOPO reversed-phase extraction behavior of rutherfordium in HCl solutions

Toyoshima, A.,¹ Kasamatsu, Y.,¹ Tsukada, K.,¹ Haba, H.,² Asai, M.,¹ Ishii, Y.,¹ Toume, H.,¹ Sato, K. T.,¹ Nishinaka, I.,¹ Nagame, Y.,¹ Goto, S.,³ Ishiyama, T.,³ Sakamaki, T.,³ Kudo, H.,³ Akiyama, K.,⁴ Oura, Y.,⁵ Kikunaga, H.,² Ooe, K.,⁶ Kuribayashi, T.,⁶ Shinohara, A.,⁶ Sueki, K.,⁴ Yokoyama, A.⁷ (¹JAEA, ²RIKEN, ³Niigata Univ., ⁴Univ. Tsukuba, ⁵Tokyo Metropolitan Univ., ⁶Osaka Univ., ⁷Kanazawa Univ.)

TOPO (trioctylphosphine oxide) reversed-phase extraction behavior of Rf in HCl solutions was investigated together with Zr and Hf to systematically study extraction behavior of Rf with organophosphorous compounds. It was found that the extraction probabilities of Rf, Zr and Hf into TOPO start to increase at 2.0 M HCl and reach to 100% at around 7.0 M HCl. The HCl-concentration range of 2.0 to 7.0 M where the extraction probabilities of the three elements into TOPO increase is lower than that of 7.0 to 8.0 M found in the previous TBP-extraction study. Thus, it seems that coordination of TOPO to the tetrachloride complexes of Rf, Zr and Hf is stronger than that of TBP.

4P14 : Adsorption of Nb, Ta and Pa on anion exchangers in diluted HF media

Kasamatsu, Y.,¹ Toyoshima, A.,¹ Toume, H.,¹ Tsukada, K.,¹ Haba, H.,² Nagame, Y.¹ (¹JAEA, ²RIKEN)

Anion-exchange behavior of Nb, Ta and Pa in HF/HNO₃ media was investigated in order to find suitable experimental conditions for the study of the Db fluoride complex formation. We measured K_d values of those elements on strong basic anion exchangers in the wide range of the fluoride ion concentration, $1 \times 10^{-6} \text{ M} < [\text{F}^-] < 2 \times 10^{-2} \text{ M}$. We discuss the formation of anionic fluoride complexes of the elements on the basis of the variation of the K_d value against [F⁻] and [NO₃]. Based on these results together with the measurement

of the equilibrium time, we propose rapid online experimental condition for the study of the Db fluoride.

4P15 : EXAFS study for complexation of Zr and Hf with TBP in HCl solution

Akiyama, K.,¹ Haba, H.,² Tsukada, K.,³ Asai, M.,³ Toyoshima, A.,³ Yaita, T.,⁴ Sueki, K.,¹ Nagame, Y.³ (¹Univ. Tsukuba, ²RIKEN, ³ASRC, JAEA, ⁴JAEA)

Recently, we reported that the order of TBP extraction for the group IV elements of the Zr, Hf, and Rf in the HCl system is $Zr > Hf \approx Rf$ using a TBP resin for the reversed phase extraction chromatography. For understanding of the difference in this extraction behavior, we study the complex structure of the Zr and Hf in the TBP resin using Extended X-ray Absorption Fine Structure (EXAFS) measurement. The observed Radial Distribution Functions (RDF) of the Zr and Hf are almost same as each other in the observed concentration range. In the session, we discuss the TBP extraction behavior of the group IV elements by the structural data deduced from EXAFS measurement of the Zr and Hf in the TBP resin together with those in the HCl solution and an anion exchange resin.

4P16 : Elucidation of ¹⁰Be accumulation mechanism to seafloor with the marine sediment.

Yoshida, T.,¹ Yamagata, T.,¹ Saito, T.,² Nagai, H.,² Matsuzaki, H.³ (¹Grad. School of Integrated Basic Sci., Nihon Univ., ²College of Humanities and Sci., Nihon Univ., ³School of Eng., Univ. of Tokyo)

Marine sediment samples (0-30 cm in depth) were collected in the Northwest Pacific Ocean, and South Pacific Ocean during KH00-3 (BO, 7 samples) and KH04-5 (SX, 8 samples) cruise of R/V Hakuho-Marui. The ¹⁰Be concentration in the marine sediment samples range between 0.9×10^9 and 6.5×10^9 atoms/g, and most of the red clay sediment in the Northwest Pacific Ocean showed uniform distribution. The ⁹Be concentration in the red clay sediment samples range between 2.3 and 2.6 ppm, which showed a value almost the same as measured ⁹Be concentration (1.6-2.3 ppm) in the Chinese loess. The ¹⁰Be concentration in the marine sediment were 20 times higher than the ¹⁰Be concentration (0.2×10^9 atoms/g) in the Chinese loess. These results were suggested that most of ¹⁰Be in the marine sediment were regarded seawater as the origin.

4P17 : Distribution of U and Th isotopes in lake sediment core-Lake Hovsgol, Mongolia

Yamamoto, M.,¹ Sakaguchi, A.,² Tomita, J.,² Kashiwaya, K.¹ (¹K-INET, Kanazawa Univ.)

Two sediment cores from Lake Hovsgol in Mongolia, with length of 81-m (HDP-04) from the central part and 7.2m (BB-04) from the Bortsog Bay, have been measured for U and Th isotopes to study their sedimentation behaviors with respect to paleoenvironmental changes. The U and Th isotopes in lake bottom sediments mainly arise in two different ways, i.e., direct inflow of clastic matter (terrestrial U or Th) and delivery from adsorption and/or adhesion onto settling particles (authigenic U or Th). Dissolved ²³⁸U concentration in the lake water is as high as

6-8 mBq/L, and its ²³⁴U/²³⁸U activity ratio exhibits distinctly high ratio of about 2. The ²³⁸U contents for long core HDP-04, with a approximate age of >1 Myr at the bottom, fluctuated by a factor of 7, ranging from 14 to 97 mBq/g, presumably reflecting the environmental changes between interglacial and glacial periods. This study highlights the potential use of authigenic and terrigenous U (Th) signatures in sediments to trace the behavior of these elements and to obtain insight into their implications for paleoclimatic variation.

4P18 : Determination of ²²⁶Ra and ²²⁸Ra in surface seawater around Sagami Bay and its vicinity

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Activity ratios of ²²⁸Ra/²²⁶Ra in surface seawaters were observed at 5 ports of Izu-Oshima, Ajiro port of Sagami bay and Umizuri park of Tokyo bay. The ²²⁸Ra/²²⁶Ra activity ratio in surface seawaters were from 0.2 to 0.7 for Izu-Oshima, from 0.2 to 0.7 for Ajiro port and from 0.6 to 0.8 for Tokyo bay. The activity ratio in surface seawaters of Izu-Oshima and Atami of Sagami bay showed lower in spring and highly in autumn, while narrow variation pattern was observed in Umizuri park of Tokyo bay. The observed variation patterns of ²²⁸Ra/²²⁶Ra in surface seawaters at Izu-oshima, Ajiro park and Umizuri park may be reflected by difference of contribution from amount of seawater far off coast with low ²²⁸Ra concentration.

4P19 : Vertical profiles of thorium isotopes with the multiple-unit large-volume *in situ* filtration system in the North Pacific

Aono T., Nakanishi T., Yamada M., Kusakabe M. (NIRS)

Thorium isotopes have been used as a tool to estimate transportation and transformation rates of particles in oceanic surface layer, as these are known to have a strong affinity to particles in seawater. It is difficult to concentrate and fractionate the particulate matters with the general filtration systems in seawater, because the concentrations of radionuclides and particles are very low in the ocean. We have developed an *in situ* filtration and concentration system. Samples were taken using the multiple-unit large-volume *in situ* filtration systems during the two cruises in the North Pacific. The concentrations of ²³⁴Th in the large (>70μm) and small (1-70μm) particulate form were 0.01-0.2 dpm/L in the depth of 200 m. The ratios of POC/²³⁴Th(μmol/dpm) in the large particles were 1-1.8 in the upper 50 m depth and less than 1 in the 100 -150 m depth. It is considered that the biological activity and particle size played a key role in controlling trends in the POC/²³⁴Th ratio in oceanic surface layer.

4P20 : High yield separation of trace plutonium from manganese crust

Sato, Y., Kinoshita, N., Yokoyama, A., Nakanishi, T. (Grad. School Nat. Sci. Tech., Kanazawa Univ.)

For a study searching for extraterrestrial Pu-244 in deep-sea manganese crust, it is essential to separate trace plutonium from kg amount of manganese crust with high

chemical yield. In the present work, chemical yield of plutonium was investigated to establish a high yield method to separate trace plutonium from manganese crust. It was found that chemical yield of Pu was improved by adopting fluoride coprecipitation procedure and by reducing Pu with sodium nitrite before anion-exchange column method. It was also confirmed that samarium hydroxide method to prepare counting source was effective.

4P21 : Determination of Pu isotopes in settling particles at femtogram levels using SF-ICP-MS with a high-efficiency sample introduction system

Zheng, J., Yamada, M. (Nakaminato Lab. Marine Radioecology, NIRS)

We report an analytical method for the determination of Pu concentration and its isotope ratio ($^{240}\text{Pu}/^{239}\text{Pu}$) for settling particle samples by SF-ICP-MS. Efforts were made to improve the sensitivity of SF-ICP-MS and reduce the $^{238}\text{U}^+$ interference for Pu analysis by combining a high-efficiency sample introduction system (APEX-Q). An extremely low detection limit of 0.07 fg Pu was achieved, which allowed the determination of Pu isotope ratio at femtogram levels. Simple anion-exchange chromatography for the separation and purification of Pu was combined with the APEX-Q/SF-ICP-MS system to determine Pu isotopes in settling particles collected in the East China Sea continental margin and in the Okinawa Trough. The obtained results supported a previous observation on the lateral transport of Pu containing particles in this continental margin, and suggested that advective lateral transport of dissolved Pu from the open ocean to the ocean margin and removal of Pu into the margin sediments by particle scavenging is a common phenomenon in the Pacific Ocean.

4P22 : $^{239,240}\text{Pu}$ inventories in marine sediments in the Pacific near Japan

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

$^{239,240}\text{Pu}$ ($T_{1/2} = 2.41 \times 10^4$ y, 6.56×10^3 y) in the environment were released from atmospheric nuclear tests carried out between 1945 and 1980. The ocean, by virtue of its large surface area, has received a major share of the fallout radionuclides. With the aim of understanding factors controlling the sinking process of Pu from water column to sediment, we measured the depth profile of Pu in sediments from the Pacific near Japan and compared the Pu inventory in the sediment column with that in the water column of the same sea area. $^{239,240}\text{Pu}$ inventory in sediment column in the Izu-Ogasawara Trench were determined to be ~ 1.7 mBq cm^{-2} , and that in the water column to be ~ 4.6 mBq cm^{-2} . This indicates that ~ 27 % of fallout Pu reached on the sea-surface has entered into the sediment in the Izu-Ogasawara Trench by the year 1994. In Japan Trench, it was found that ~ 50 % of fallout Pu entered into the sediment by the year of 1994.

4P23 : Depth distribution of ^{241}Am in water columns of the Eastern Indian Ocean and its adjacent seas

Izumi, T., Nakanishi, T. (Grad School Nat. Sci. Tech., Kanazawa Univ.)

^{241}Am ($T_{1/2} = 432.2$ y) in seawater is produced from dissolved ^{241}Pu ($T_{1/2} = 14.35$ y) by β^- decay. With the aim of understanding the sinking behavior of Am in seawater, depth distributions of ^{241}Am were determined in water columns in the Eastern Indian Ocean and its adjacent seas. It was found that ^{241}Am concentration increased from surface to 1000 m and showed constant from 1000 m to the bottom. The depth distribution pattern of ^{241}Am concentration can be explained by a model that ^{241}Am produced from ^{241}Pu between surface and 1000 m attached to sinking materials which goes to bottom at almost constant velocity.

4P24 : Radioactivity of uranium-series and thorium-series nuclides in Tamagawa hot-spring water, Akita Prefecture

Saito, T.,¹ Yamagata, T.,² Yoshida, T.,² Tsukahara H.,² Nagai, H.¹ (¹College of Humanities & Sci., Nihon Univ., ²Grad. School of Integrated Basic Sci., Nihon Univ.)

The concentrations of uranium and the progenies (^{238}U , ^{234}U , ^{230}Th and ^{226}Ra) and thorium and the progenies (^{232}Th , ^{228}Ra and ^{228}Th) in Tamagawa hot-spring water were investigated. Activity of ^{238}U , ^{234}U , ^{232}Th , ^{230}Th and ^{228}Th were measured by α -ray spectrometry, and the ^{226}Ra , ^{228}Ra and ^{228}Th were analyzed by a well-type HPGe γ -ray spectrometer for the 351, 911 and 239 keV γ -ray from ^{214}Pb , ^{228}Ac and ^{212}Pb , respectively, each being in radioactive equilibrium with precursors. The ^{238}U , ^{234}U and ^{228}Th concentrations were virtually constant, and the ^{232}Th , ^{230}Th , ^{228}Ra and ^{226}Ra concentration were high tendencies at the summer season.

4P25 : Behaviors of uranium series nuclides in borehole groundwaters at Kanamaru, Yamagata Prefecture

Kanai, Y., Seki, Y., Okuzawa, K. and Kamioka, H. (AIST, Geological Survey of Japan)

In order to elucidate the movement-retention behavior of uranium in groundwater, the uranium series nuclides such as U-238, U-234 and Th-230 in the borehole groundwaters at Kanamaru, Niigata Prefecture, were studied. Water samples were collected from Br1, Br3-3 and Br4 in 2002 and 2005 and activities were measured by alpha spectrometry after separation and purification of U and Th by anion-exchange resins and electrodeposition. Although most of the samples showed the activity ratios of U-234/U-238 > 1 and Th-230/U-234 < 1, some showed those of U-234/U-238 > 1 and Th-230/U-234 > 1 in earlier survey (2002). This may suggest the existence of suspended and/or colloidal particles in the upper surface groundwater.

4P26 : Environmental tritium dynamics model in a small drainage basin

Hirose, A.,¹ Sugihara, S.,² Momoshima, N.,³ Maeda, Y.,² Satake, H.⁴ (¹Grad. School of Sci., Kyushu Univ., ²Fac. Sci., Kyushu Univ., ³Radioisotope Center, Kyushu Univ., ⁴Fac. Sci., Toyama Univ.)

Environmental water, such as rain water and the river water, mutually has close relations. The tritium can be an important index in thinking about this water circulation. So the concentrations of the tritium were measured for

environmental water samples. We researched in the river and the lakes and marshes water of the whole country to understand the background concentration of a present tritium. The concentrations of the tritium is in the range between 0.42 ± 0.027 and 2.66 ± 0.041 Bq/L and its average is 1.16 ± 0.040 Bq/L. Moreover, we estimated the underground water residence time by the compartment model in the Umi river, Fukuoka Prefecture.

4P27 : $^{230}\text{Th}/^{234}\text{U}$ activity ratio in volcanic products from Kozu-shima and Nijima

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

With 45 volcanic rocks erupted recently from 6 volcanoes located along Izu-Mariana island-arc, Japan, ^{234}U is reported to be in radioactive equilibrium with ^{238}U . Observation was made with rhyolitic volcanic rocks erupted from Izu-Mariana island-arc volcanoes for activity ratios of $^{230}\text{Th}/^{234}\text{U}$ in order to discuss on the radioactive disequilibrium between ^{234}U and ^{230}Th in the latest stage of magmatic evolution. The $^{230}\text{Th}/^{234}\text{U}$ activity ratios of 7 rhyolitic products from 2 volcanoes of Izu-Mariana island-arc, Kozu-shima and Nijima, indicated that ^{230}Th was in radioactive equilibrium with ^{234}U in the erupting magmas.

4P28 : Estimation of effective dose from Rn emanating from "the minus ion" effect wallpaper

Yoshizawa, Y.,¹ Minowa, H.,¹ Morita-Murase, Y.,² Furuta, E.³ (Jikei Univ.,² Kyoritsu Pharm. Univ.,³ Ochanomizu Univ.)

We have examined the wall papers which declared "the minus ion" effect to estimate external and internal exposure dose from them. Results of gamma-ray spectrometry revealed that they contain 0.03 to 0.35 Bq·g⁻¹ of Th-series nuclides, ^{208}Tl , ^{212}Pb , ^{212}Bi and ^{228}Ac , and U-series one, ^{214}Pb . Distributions of radioactive nuclides in the samples were measured using an imaging plate and a FLA-2000 (Fuji Photo Film). The radiation doses from the printed side of the wall papers were 5 to 15 times higher than that of the back side. The ^{222}Rn concentrations emanating from the wall papers in a sealed container of 50 liter were measured using the PICO-RAD radon detectors. One wall paper showed two to five times higher than the background value.

4P29 : High resolution simultaneous measurements of airborne radionuclides

Abe, T., Yamaguchi, Y., Tanaka, K., Komura, K. (LLRL Kanazawa Univ.)

High resolution (2-3 hrs) simultaneous measurements of airborne radionuclides, ^{212}Pb , ^{210}Pb and ^7Be , have been performed by using extremely low background Ge detectors at Ogoya Underground Laboratory. We have measured above radionuclides at three monitoring points viz, 1) Low Level Radioactivity Laboratory (LLRL) Kanazawa University, 2) Shishiku Plateau (640 m MSL) located about 8 km from LLRL to investigate vertical difference of activity levels, and 3) Hegura Island (10 m MSL) located about 50 km from Noto Peninsula in the Sea of Japan to evaluate the influences of Asian continent or mainland of Japan on the variation to the activity levels. Variations of short-lived ^{212}Pb concentration showed noticeable time lags between at LLRL

and at Shishiku Plateau. These time lags might be caused by change of height of a planetary boundary layer. On the contrary, variations of long-lived ^{210}Pb and ^7Be showed simultaneity at three locations because of homogeneity of these concentrations all over the area.

4P30 : High resolution simultaneous measurements of airborne radionuclides in the pan-Japan sea area

Yamaguchi, Y.,^{1,2} Abe, T.,¹ Murata, Y.,¹ M, Manikandan, N.,¹ Tanaka, K.,¹ Komura, K.¹ (¹LLRL, Inst. Nat. Env. Tech., Kanazawa Univ., ²Radioisotope Center, Osaka City Univ.)

By the use of ultra low background Ge detectors at Ogoya Underground Laboratory (OUL), it became possible to detect extremely low levels of environmental radionuclides. In this study, we tried to measure high resolution simultaneous measurements of airborne radionuclides at three monitoring points, i.e., 1) Low Level Radioactivity Laboratory (LLRL 40m asl) in Nomi City as the regular monitoring point, 2) Hegura Island located 50 km from Noto Peninsula in the Sea of Japan to investigate the influence of Asian continent or mainland of Japan, and 3) Shishiku Plateau (640m asl) located about 8 km from LLRL to know vertical difference. Pb-210 and Be-7 were measured nondestructively by ultra low background gamma spectrometry at OUL, Po-210 by alpha spectrometry using Si detectors after the chemical treatment. Various interesting results on the concentrations and variation patterns of airborne radionuclides were obtained, particularly, during drastic meteorological changes such as the passage of typhoon, snow fall and so on. We have been analyzing the influence of the arrival of yellow sand occurred in this spring.

4P31 : Characterization of fallout deposition materials during 1993-2005 at Tokai-mura by radioanalytical methods

Namioka, H.,¹ Nagao, S.,¹ Irino, T.,¹ Ueno, T.,² Aramaki, T.,³ Shibata, Y.³ (¹Hokkaido Univ., ²JAEA., ³NIES)

Characteristics of fallout deposition materials collected during 1993-2005 at Tokai-mura were carried out by radioanalytical methods. The deposition samples were taken by a basin (surface area of 0.5m²) during one month, and dried-up, powdered by an agate mortar. The $^{14}\text{C}/^{12}\text{C}$ ratios were measured for the samples in March of 1996, 1998, 2000, 2002 and 2004 by accelerator mass spectrometry at the NIES. Deposition fluxes of organic carbon ranged from 0.11 to 2.25 g/m²/month, showed two peaks in spring and autumn every each year, and almost showed maximum in spring. The $\Delta^{14}\text{C}$ values were from -80.2 to -5.5‰. When the $\Delta^{14}\text{C}$ values were plotted as a function of deposition flux of organic carbon, it appears the correlation between $\Delta^{14}\text{C}$ and TOC flux. These values suggest that organic carbon deposited in each year may be mixed with newly formed and old organic carbon.

4P32 : Seasonal variations of the source of aerosol particles by using backtrajectories and principal component analysis

Morinaga, I., Sugihara, S., Maeda, Y. (Kyushu Univ.)

Atmospheric aerosol is important component in

considering the effect on human beings, climates and the reaction processes in atmosphere. Therefore, many researchers are interested in its movement in atmosphere. The purpose of this work is to consider the seasonal variations of the source of aerosol particles by the principal component analysis and the backtrajectories. The samples were collected by a container routinely every month at Fukuoka city (the rooftop of Radioisotope Center, Kyushu University) from Jan. 1995 to Jun. 2006. The concentrations of radionuclides and elements were determined by a gamma-ray spectrometry and a neutron activation analysis. As a result of the principle component analysis and the backtrajectories, we found some sources which contributed to the components of the aerosol particles at Fukuoka City.

4P33 : Development of detection method for individual environmental particles containing alpha radioactive nuclides

Esaka, K. T., Yasuda, K., Esaka, F., Magara, M., Sakurai, S., Usuda, S., Nakayama, S. (JAEA)

Artificial radioactive nuclides have been emitted from various sources and have fallen on the surface of the earth as fine particles. Although the characterization of the individual fallout particles is very important, their analysis is difficult. The purpose of this study is to develop a new detection method for individual objective particles containing radioactive nuclides in the environment. The soil or sediment sample was confined in a plastic film and the locations of objective particles were identified with alpha tracks created in a solid-state detectors (BARYOTRAK, Fukuvi Chemical, Ltd) stuck to the both sides of the plastic film. A piece of the film containing the objective particle was cut with a nitrogen laser for following individual particle analysis. This procedure allowed us to detect the objective particle from innumerable number of particles in the environment and characterize the individual particles.

4P34 : Determination of radionuclides contaminated in chemical reagents

Inoue, M., Komura, K. (LLRL, Kanazawa Univ.)

In order to examine reagent blanks of radionuclides (^7Be , ^{60}Co , ^{134}Cs , ^{137}Cs , ^{210}Pb , ^{226}Ra , ^{228}Ra and ^{234}Th), we applied extremely low-background gamma-spectrometry to Ba, Pb and other reagents used for coprecipitation methods. While blank levels of ^{226}Ra and ^{228}Ra in Ba reagents and ^{210}Pb in Pb reagents were high and varied widely (e.g., ^{226}Ra , 6-90 mBq/g-Ba), regardless of their chemical purity, they were notably lower in barite mineral (BaSO_4) (0.7-1.5 mBq/g-Ba for ^{226}Ra) and old Pb (from the Kanazawa Castle, the Edo period). In addition, the negligible ^{134}Cs and ^{137}Cs were detected in CsCl reagents. By using least contaminated carrier reagents together, the coprecipitation method has enabled simultaneous measurement of low-level activities of these nuclides in sea/environmental water samples.

4P35 : On-line monitoring of radioactive airborne dust using high-speed pulse interval analysis with multi-input channels

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Monitoring, ²ZCOSMOS Co., Ltd., ³Fac. Sci., Niigata Univ.)

A portable system for high-speed Time Interval Analysis (TIA) method was constructed. The system installed with an original time counting board could record pulse incident time of 1 MHz precisely. During the measurement, calculation of time intervals for all the pulses and on-line display of the results were performed simultaneously. Using this system, the correlated decay events of ^{214}Bi to ^{210}Po in the airborne dust collected on the filter paper were counted, together with a dual-phosphor type alpha/beta detector. A rapidly detective technique of artificial radioactivity under the variations of naturally occurring radioactivities was discussed.

4P36 : Instrumental development for measurement of elements distribution on the surface of smear samples

Yasuda, K., Magara, M., Sakurai, S., Usuda, S., Shinohara, N. (JAEA)

In order to analyze the isotope ratios of the ultra trace amount nuclear materials in smear samples efficiently, evaluating the distribution of the impurity elements that interfere with isotope ratio measurement beforehand becomes important. Therefore, an instrument to measure the elemental distribution on the surface of the smear sample by X-ray fluorescence was developed. For analyzing of a 10×10 cm cotton smear sample, we found linearity between lead concentration and signal intensity within the range of 30 – 1000 ng/pixel when the sample was measured for 6 hours with the resolution of 5×7 mm. The detection sensitivity is sufficient, since more than 10 μg of lead interferes with isotope ratio measurement if ICP-MS is applied.

4P37 : Plutonium in the Nishiyama area of Nagasaki city

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After the detonation of Nagasaki atomic bomb, plutonium with 'black rain' was precipitated at the Nishiyama area. The source of plutonium in sediments and soils collected at the Nishiyama area was characterized by their $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio and their concentrations of $^{239+240}\text{Pu}$ and ^{137}Cs . The distribution of $^{239+240}\text{Pu}$ and ^{137}Cs showed a peak at the depth nearby 440 cm, where $^{240}\text{Pu}/^{239}\text{Pu}$ ratios were about 0.028. These suggested the plutonium isotopes and ^{137}Cs released from Nagasaki atomic bomb were deposited in the layers. The higher ratios were detected in the sediments at the depth nearby 370 cm with a peak of ^{137}Cs concentration, which showed the plutonium released from nuclear tests was mixed with the plutonium released from the atomic bomb. The concentrations and the ratios were kept constant in the sediments at the depth from 350cm to the surface. The ratios were comparable to the values in the soils collected around the reservoir. This means that the soils around the reservoir have been eroded and have flown into the reservoir constantly.

4P38 : Interference due to Sm-X-rays to the determination of ultra-low-level ^{152}Eu separated

from a sample exposed to Nagasaki atomic-bomb

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

4P39 : Radiation dose due to neutron-induced residual radioactivities by the atomic bombs in Hiroshima and Nagasaki

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Radiation dose at 1 m above the ground was estimated due to neutron-induced radioactivities by the atomic bombings in Hiroshima and Nagasaki. The dose rate at the hypocenter at 1 min after the bombing was 600 and 400 cGy h⁻¹ in Hiroshima and Nagasaki, respectively, and rapidly decreased with the decay of the main induced radioactivities of relatively short half-lives: ^{28}Al (2.24 min), ^{56}Mn (2.58 h) and ^{24}Na (15.0 h). The infinite cumulative dose after the bombing was 120 and 57 cGy at the hypocenter of Hiroshima and Nagasaki, respectively, and also sharply decreased with the distance from the hypocenter. Considering the results obtained so far, the people who entered into the area less than 1 km from the hypocenter up to 1 week after the bombing could receive external dose over 1 cGy from neutron-induced radioactivities.

4P40 : Concentrations of ^{99}Tc and ^{137}Cs in edible kelps and sea urchin ovaries from the northern part of Japan

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The first commercial nuclear fuel reprocessing plant in Japan is located in Rokkasho Village, Aomori Prefecture, and is now under the active test using actual spent nuclear fuels. Since the plant will release small amount of ^{99}Tc to the ocean, the concentration of ^{99}Tc in seawater nearby the plant will possibly increase in future. Since it is important to get background concentration levels of ^{99}Tc and ^{137}Cs in marine products from the ocean nearby the plant for assessing the effect to the concentration levels by the plant, the nuclides in

edible kelps and sea urchin ovaries from the northern part of Japan (Aomori, Hokkaido and Iwate) were observed. Concentrations of ^{99}Tc in the kelps were lower than those reported for other non-edible seaweeds (*Sargassum thunbergii*) in Aomori, and the same level to the reported value of *Hizikia fusiformis* in Kyushu. ^{99}Tc concentrations in the urchin ovaries were lower than the detection limit of our method, and concentrations of ^{137}Cs in them were similar to these in the kelps. It suggested that ^{99}Tc is not concentrated in sea urchin ovaries. This work was supported by a grant from Aomori prefecture, Japan.

4P41 : Method for measurement of ^{129}I in vegetation samples by using AMS – Target preparation –

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We investigate three methods for collecting iodine in vegetation samples in order to prepare AgI target for analysis of ^{129}I by using AMS. The methods are (1) dry combustion method, (2) oxygen bomb method and (3) alkaline leaching method. We evaluate blank level and recovery of iodine for each method.

4P42 : Depth profile of I-129 concentrations in soil determined by AMS

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Iodine-129 is one of the most important radionuclides in environmental sciences and geochemistry. In this study, we have developed an appropriate separation procedure of iodine from environmental samples such as soil for determining $^{129}\text{I}/^{127}\text{I}$ ratio by AMS (accelerator mass spectrometry). Concentrations of stable iodine were analyzed by ICP-MS. Using these methods we have analyzed soil samples collected from difference places in Japan. We also studied the depth profile of both ^{129}I and stable iodine concentrations. The detection limit of $^{129}\text{I}/^{127}\text{I}$ using this method was about 2×10^{-11} , which is about 2 orders of magnitude better than that obtained by neutron activation analysis (NAA). Concentration of ^{129}I decreased significantly with the depth, i.e. from 1.7×10^{-3} Bq/Kg (surface) to 8.1×10^{-6} Bq/Kg (at the depth of 236 cm) in field soil samples collected from Tsukuba. In case of stable iodine, the concentration was around 30 ppm and not so much changed with the depth.

4P43 : Relationships between the long-term mobility of Sr-90 in ploughed soil of upland field and soil properties

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Though more than 25 years have elapsed since the latest atmospheric nuclear test, long-lived artificial radionuclides, such as ^{137}Cs and ^{90}Sr , still remain in the soil. We evaluated the mobility of ^{90}Sr in ploughed upland soil, which affects the residual amount in the soil and plant uptake on the basis of long-term monitoring data. Soil was taken annually from 1961 to 1995 from 8 agricultural fields, and the concentration of exchangeable ^{90}Sr in soil was determined. The

concentration of exchangeable ^{90}Sr in soil decreased exponentially with time. The environmental factor responsible for the decrease of ^{90}Sr , λ_e , was determined by the exponential fit of the exchangeable ^{90}Sr in ploughed soil vs. year followed by subtraction of the physical decay constant. The main factor controlling λ_e , the long-term mobility of ^{90}Sr from ploughed soil, was the cation exchange capacity of soil. It was indicated that the entrapment of ^{90}Sr on a cation exchange site retards the downward migration and wheat uptake of ^{90}Sr from ploughed soil.

4P44 : Trace analysis of uranium, thorium, lead and lanthanoids in environmental samples - Examination of condition of anion-exchange separation (II) -

Miyamoto, Y., Saito-Kokubu, Y., Sakurai, S., Usuda, S., (JAEA)

The authors investigated separation scheme of trace amounts of U, Th, Pb and lanthanoids using an anion-exchange column to analyse the abundance and isotopic ratio of those elements in environmental samples. For reducing the procedural blank, the anion-exchange separation of the elements from an ICP-MS standard solution was carried out with high-pure or easy-to-purified reagents in a clean room of CLEAR at JAEA. The mixture of 90%-ethanol and 0.001M-oxalic acid with various concentration of HCl was used for the separation of Th and lanthanoids. It was found that the concentration of HCl should be lower than 0.01M in the ethanol-oxalic acid mixed-media to separate lanthanoids and Th. Thirty % - 40% of total amounts of lanthanoids was retained in the column due to the formation of precipitation of lanthanoid oxalate. By using the eluant containing lower concentration of ethanol, the formation of the precipitation can be suppressed, and the recovery of lanthanoids can be increased. The results of this experiment will be also presented.

4P45 : Formation of radioactive aerosol in electron linear accelerator irradiation room

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We measured the particle size distribution and concentration of non-radioactive and radioactive aerosols formed by electron irradiation to air in order to reveal characteristics of radioactive aerosols produced in the rooms of operating accelerators. In this work, an irradiation chamber was installed in accelerator target room. An aerosol-free sample air was introduced to the irradiation chamber. We used diffusion batteries, a low pressure impactor and a scanning mobility particle sizer to measure aerosol size distributions. The operation of accelerators produces both of the non-radioactive and radioactive aerosols. The non-radioactive particle size was found to be ca.110 nm and radioactive one was ca.180nm in the irradiation room air. It is suggested, similarly to aerosols in proton accelerators, that the radioactive aerosol particle is formed by attachment of a radioactive atom produced through nuclear reactions in air to the radiation-induced non-radioactive aerosol particle.