P41 Local Fields in ZnO Codoped with Co and Mn —a $^{57}$Co Emission Mössbauer Spectroscopic Study—
MORIICHI, R.1, KANO, Y.1, SATO, W.1,2
We measured internal fields in 6.25 at.% Co- and 6.25 at.% Mn-codoped ZnO (CMZO) and 11 at.% Co-doped ZnO (CZO) synthesized by a solid state reaction by means of $^{57}$Co emission Mössbauer spectroscopy. In CMZO, two doublets were observed in the spectrum at room temperature: one (D1) is assignable to a $^{57}$Fe$^{2+}$ ($\rightarrow$ $^{57}$Co) substituting for a defect-free Zn site in ZnO, and the other to $^{57}$Fe$^{3+}$ ($\rightarrow$ $^{57}$Co) in substitutional Zn sites adjacent to a Zn vacancy. However, below 100 K, the spectrum cannot be fitted only with two doublets, so we assumed the emergence of an additional magnetic component (M1). We found that the relative intensity of D1 decreases and that of M1 increases with decreasing temperature. In addition, we carried out magnetization measurements for CMZO, CZO and 6 at.% Mn-doped ZnO (MZO), and found that only CMZO shows slightly different trend from that expected for paramagnetism. The results of the $^{57}$Co emission Mössbauer spectroscopy and magnetization measurements may imply that codoping Co and Mn in ZnO can realize the emergence of dilute magnetism.

P42 Effect of molecular structure on the muon transfer process in benzene and cyclohexane
INAGAKI, M.1, NINOMIYA, K.1, KUDO, T.1, TERADA, K.1, SATO, A.1, KAWASHIMA, Y.2, TOMONO, D.2, SHINOHARA, A.1
(1Grad. Sch. Sci., Osaka Univ., 2RCNP, Osaka Univ.)
Formation processes of muonic atoms are influenced by the chemical environments of muon-capturing atoms, as it is called chemical effect. Previously, we reported that the chemical effect on the muon transfer processes in benzene and cyclohexane in the gas phase was smaller than the detection limit. In this study, to investigate the chemical effect on the muon transfer processes in the liquid phase, muon irradiation experiments for liquid mixtures of benzene or cyclohexane and carbon tetrachloride were carried out. The experiments were conducted at the MuSIC-M1 beamline of the Research Center for Nuclear Physics (RCNP) in Osaka University. Muonic X-rays emitted after muon-capture by carbon or chlorine atoms were measured with high purity germanium detectors. The muon capture probabilities of carbon and chlorine atoms were determined from the muonic X-ray spectra. The model formula for the muon capture was set up, and the muon transfer rate to carbon atoms in each sample was obtained using the muon capture probabilities. The result suggested that the muon transfer rate to carbon atoms of benzene molecules are higher than that to carbon atoms of cyclohexane molecules.
P43  Observation of local structures in perovskite oxide SrTiO₃ with the $^{111}$Cd probe
KOMATSUDA, S.¹, SATO, W.², OHKUBO, Y³.

Strontium titanate (SrTiO₃) doped with metal ions as impurity donors exhibits a wide variety of electric properties. For a practical use of SrTiO₃ as a conduction-controlling device, it is of great importance to study the lattice site location of doped impurities and their surroundings. For that purpose, the time-differential perturbed angular correlation method (TDPAC) is very suited because it can directly provide atomic-level information of impurity atoms. From this point of view, we have applied the TDPAC method to a study of SrTiO₃ using the $^{111}$In($\rightarrow^{111}$Cd) probe. Strontium titanate sample doped with the $^{111}$In($\rightarrow^{111}$Cd) was synthesized by a solid-state reaction in air at 1473 K. For the PAC spectrum obtained at room temperature, two electric field gradients were observed, characterized by the quadrupole frequencies and its fractions $\omega_Q = 48.9(1)$ Mrad/s, $f_1 = 68(3)\%$, and $\omega_Q^2 = 52.1(2)$ Mrad/s, $f_2 = 36(3)\%$, respectively. In the presentation, based on the further investigation of TDPAC spectra for SrTiO₃ measured at 500–1000 K, we discuss the site occupation of the $^{111}$In($\rightarrow^{111}$Cd) probe and their local structures in perovskite oxide SrTiO₃.

P44  Hydrogen-Induced Defects in Palladium Observed by Means of Gamma-Ray Nuclear Spectroscopies
FURUMOTO, M.¹, SHIMIZU, H.¹, OHKUBO, Y.², SATO, W.¹,³

Palladium (Pd) can dissolve a large amount of hydrogen atoms in the lattice interstitial sites and is expected to be applied as a hydrogen storage material. However, it is widely known that defects formation induced by hydrogen absorption may decrease its storage performance. Thus, application of Pd to hydrogen storage metal still has problems in terms of tolerance. Therefore, in order to obtain more detailed information on the hydrogen storage performance of Pd, it is important to investigate the physical state of lattice defects caused by the introduction of hydrogen. In this study, behavior of lattice defects in Pd associated with hydrogen absorption was observed by two different nuclear spectroscopies. Positron annihilation lifetime spectroscopy (PALS) applied to electrochemically hydrogen-introduced Pd suggested existence of lattice defects. Perturbed angler correlation (PAC) measurements using the $^{111}$Ag probe revealed the presence of electric field gradient at the probe site in the electrochemically hydrogen-introduced Pd. In the poster presentation, we evaluate the hydrogen-induced lattice defects in Pd based on the results of the nuclear spectroscopic methods.
P45 Production method development of $^{67}$Cu using thermoseparation for cancer therapy.  
OHTA, A.1,2, KAWABATA, M.1,2, MOTOISHI, S.1,2, SAEKI, H.1,2, HASHIMOTO, K2., TSUKADA, K. 2,3, HATSUKAWA, Y. 2, NAGAI, Y. 2  
(Chiyoda Technol Corporation1, National Institutes for Quantum and Radiological Science and Technology2, Japan Atomic Energy Agency3)

$^{67}$Cu is a promising RI for both cancer therapy and diagnosis in nuclear medicine, yet the large scale production has not been achieved to satisfy the clinical demand. High quality $^{67}$Cu can be produced via the $^{68}$Zn(n,x)$^{67}$Cu reaction ($x=n',p,d$) from zinc target where projecting fast neutrons are generated in carbon converter by stopping 40 MeV deuterons from an accelerator. Separation of $^{67}$Cu from $^{68}$Zn were previously established1, however it may require a longer hours to obtain a final product from a large volume of zinc target. Concentration of a large quantity of zinc was therefore investigated by thermoseparation2 using a difference in vapor pressure between zinc and copper. The method also serves the removal any impurity copper in the zinc target before irradiation.

Natural zinc metal that was irradiated by neutrons at CYRIC Accelerator (Tohoku University) or Tandem Accelerator (Japan Atomic Energy Agency) was heated under reduced pressure. The bulk of zinc was sublimated, moved and deposited at lower temperature area. The gamma-rays from $^{67}$Cu and $^{69m}$Zn within the target were measured with CZT (CdZnTe) semiconductor detector. The poster discusses the results on separation efficiencies and the behavior of both tracer elements while they were heated.

Reference
2) Lu Zhan et. al., Separation and Purification Technology 68, 397 (2009)

P46 Development of a Rn-At generator system using radon gas enclosed within a syringe  
KAWASAKI, K.1, SHIN, Y.1, AOI, K.2, WASHIYAMA, K.3, NISHINAKA, I.4, HABA, H.5  
YANOU, S.5 YOKOYAMA, A2  
(1Graduate School of Natural Science and Technology, Kanazawa University., 2Institute and College of Science and Engineering, Kanazawa University., 3Fukushima Medical University., 4Tokai Quantum Beam Science Center, QST., 5RIKEN Nishina Center.)

The radionuclide of $^{211}$At gathers much attention as a nuclide useful for isotope treatment. Its chemical species are explored and the solvent extraction of At is investigated for that purpose. However, its half-life is as short as 7.2 hours and it is difficult to use it in a place remote from an accelerator. Milking of $^{211}$Rn (of 14.6 hours in half-life), the parent nuclide of $^{211}$At, makes it possible to use it at a remote location. In this study, $^{211}$Rn gas was stored in a sealed syringe form a dissolved target. $^{211}$At produced $^{211}$Rn gas with nitric acid or hydrochloric acid. Beside we investigated on the extraction and back extraction of $^{211}$At using ionic liquid as solvent and crown ether as extractant. As a result, we succeeded in holding $^{211}$Rn gas in a syringe and recovering $^{211}$At from $^{211}$Rn gas. The $^{211}$At extraction was performed at an extraction rate of about 90% with 0.1M nitric acid. The back extraction of At was performed with sodium hydroxide and potassium hydroxide at their higher concentrations than 1M. From the results, we conclude that this method is promising as a generator system.
P47  Syntheses and stability evaluation of alkaline earth metal complexes for nuclear medicine
NAGATA, K.1, YAJIMA, T.2, KASAMATSU, Y.3, SHINOHARA, A.3, YOSHIMURA, T.1
(1Institute for Radiation sciences, Osaka Univ., 2Materials and Bioengineering, Kansai Univ., 3
Graduate School of science, Osaka Univ.)

Radium-223(223Ra) has a great potential for targeted alpha-ray therapy due to multiple alpha decay in the decay chain. The maximum value of the stability constants log$K_{ML}$ reported in the known Ra(II) complexes still remains at $\sim$10, and the log$K_{ML}$ value is insufficient to apply in nuclear therapy. We have tried to synthesize a stable Ra complex in vivo to obtain new cancer drugs applicable to various kinds of tumors. In the present study, we used macrocyclic ligands, L1 and L2, that have two 6-methyl picolinate arms and a diaza-18-crown-6-ether framework.

The alkaline earth metal (Ca, Sr, Ba) complexes were synthesized by the reactions of L1 and L2 with the metal ions, respectively. The stability constants in the Ca(II), Sr(II), and Ba(II) complexes with L1 and L2 were determined by pH-potentiometric titration in 0.1 M KNO3(aq). In all the metal ions studied, the log$K_{ML}$ values in the L1 complexes are high compared with the values in the L2 complexes. The log$K_{ML}$ values follow the order: Ba(II) > Sr(II) > Ca(II) for both the L1 and L2 complexes. It has been found that L1 can provide radionuclide ($^{85}$Sr, $^{133}$Ba and $^{226}$Ra)-chelate complexes. The reactions detected by the radio TLC method with $\gamma$ ray spectroscopy. The complex formation was quantitative in both the cases using $^{85}$Sr, $^{133}$Ba and $^{226}$Ra.
Investigation on solvent extraction of astatine for $^{211}$Rn-$^{211}$At generator — Comparison to the $^{131}$I behavior and investigation on the effect of oxidizing agents in extraction

SHIN, Y$^1$, KAWASAKI, K$^1$, AOI, K$^2$, YOKOYAMA, A$^2$, WASHIYAMA, K$^3$, NISHINAKA, I$^4$, YANOU, S$^5$, HABA, H$^5$.

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$^{211}$At is a promising nuclide among various $\alpha$-emitters for targeted $\alpha$ therapy. It has high $\alpha$-particle energy and the emission probability of $\alpha$-ray is about 100%. However, $^{211}$At is produced by an accelerator and the half-life of $^{211}$At is only 7.2 hour. It is too short to deliver to remote areas from accelerator facilities. In order to use astatine in wider ranges we are developing a $^{211}$Rn-$^{211}$At generator. In a previous study, $^{211}$At produced in decaying of $^{211}$Rn in dodecane solution can be back-extracted into 90% ethanol aqueous solution with N-bromosuccinimide (NBS), while the mechanism of astatine extraction with NBS is not known. In this study we assumed that Br$_2$ generated from NBS oxidizes astatine. We used NCS and NIS instead of NBS to investigate the effect of redox potential. In addition, we compared the back-extraction ratios of $^{211}$At and $^{131}$I by using bromine water as an oxidizing agent. As a result, the order of back extraction ratio is NBS > NCS > NIS. Comparing $^{211}$At and $^{131}$I, the back-extraction ratio of $^{211}$At increases with addition of bromine, but that of $^{131}$I does not increase. It shows that $^{211}$At is oxidized into species which is easily extracted.
P51 Synthesis of an astatinated amino acid derivatives via silicon-astatine exchange reaction
WATANABE, S.1, ANWAR-UL, A.M.1,2, NISHINAKA, I.1, SASAKI, I.1,3, OHSHIMA, Y.1, YAMADA K.1, ISHIOKA, N.S.1
(1QST Takasaki, 2NINMAS, BAEC, 3Graduate School of Science and Technology, Gunma Univ.)
Astatine-211 (211At) is a promising alpha-emitting radionuclide for the targeted alpha therapy (TAT) and tin-astatine exchange reaction has been used for the synthesis of astatinated compounds. However, organotin compound is readily decomposed under high temperature and acidic condition, which makes synthetic routes of precursor complicated. In addition, toxicity of the organotin compounds is a potential cause of concern for future clinical applications. We therefore focused on silicon-astatine exchange reaction. In the present study, 4-[211At]astato-L-phenylalanine was synthesized via the electrophilic desilylation. HPLC analysis revealed that a strong radioactive peak was observed at 16.8 min. of retention time, which was identical to that of corresponding radiiodinated phenylalanine (retention time: 15.8 min.). The astatinated compound was time-dependently accumulated into LS180, colon adenocarcinoma cell, and the uptake was inhibited in the presence of some amino acid derivatives. These results clearly indicated that 4-[211At]astato-L-phenylalanine was successfully synthesized in this study. Radioc hemical yield and purity were 64-75% and over 99%, respectively. We therefore make a conclusion that the silicon-astatine exchange reaction is useful to synthesize astatinated amino acid derivatives and peptides.

P52 Study on synthesis of Y@C60 by nuclear recoil
ITO, Y.1, AKIYAMA, K1, KUBUKI, S.1, HATSUKAWA, Y.2
(1Tokyo Metropolitan Univ, 2QST.)
We have tried to synthesis of Radioactive Endohedral Metallofullerenes (REMFs) using the nuclear recoil induced by nuclear reaction. This method has some advantages from the viewpoint of a small amount of by-products and selective production of target REMFs. In the previous study, we reported the synthesis of 85Sr@C60 using the nuclear recoil 86Sr(n, 2n)85Sr reaction with 14 MeV fast neutron induced by DT reaction. In this study, we will report the synthesis of 88Y@C60 by nuclear recoil of 89Y(n, 2n)88Y reaction with 30 MeV and 50 MeV fast neutron induced by DT reaction.
Irradiated samples were dissolved into CS2 and filtered to remove a large amount of C60. After dryness of the filter, aniline and HClaq was passed through the filter, sequentially. The γ ray from extracted samples was measured by Ge detector.
As a result, the radioactivity of 88Y observed in the aniline solutions derived from 88Y@C60 for examined in this work was estimated to be several percents in the total radioactivity of each irradiated sample.
P53  Preparation of Carrier-free $^{139}$Ce Tracer for Endohedral Metallofullerene Study
AMEKURA, K. 1, AKIYAMA, K. 1, 2, KUBUKI, S. 1, HABA, H 2
(1Tokyo Metropolitan Univ., 2RIKEN, Nishina center.)

We have studied the electronic state for a series of light lanthanide (Ln) metallofullerene, such as Ln@C$_{82}$, by analyzing the small difference in the HPLC retention time. For such precise analysis, the radiochemical method is one of the most effective approach because this method allows the simultaneous analysis of multiple elements. For further studies, it is necessary to analyze from the difference between these retention times monitored by gamma ray of reference carrier-free Ln@C$_{82}$ and by UV absorption because some heavy lanthanide, such as Er and Dy, only produce nuclides with short half-life. In this study, we report the chemical separation of $^{139}$Ce used as a carrier-free reference material from a target material of La and the result of metallofullerene preparation using this carrier-free $^{139}$Ce.

$^{139}$Ce and a target material of La were separated by the solvent extraction method using K$_2$Cr$_2$O$_7$ as an oxidant. The amount of $^{139}$Ce was determined by gamma-ray measurement and the residual amount of Cr and La were estimated to be 2.5% and 0.03%, respectively, from UV/vis absorption analysis. More than 90% of $^{139}$Ce were finally obtained and also EMF is found to be successfully prepared using this carrier-free $^{139}$Ce.

P54  A study on the sedimentary environment of Tokyo-bay sediments under hypoxia.
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Hypoxia is a big problem particularly in inner bay such as Tokyo Bay. To understand and solve the problem, we attempted to estimate the redox environment of Tokyo-bay sediments under hypoxia, assuming that the effect of past hypoxia could be preserved as a difference in concentrations and/or chemical states of elements in the sediments.

We collected sediment cores off the coast of Yokohama (flat seafloor) and Makuhari (flat seafloor and dredged trench) in Tokyo Bay. The samples were sliced every 2 cm and analyzed by instrumental neutron activation analysis. As a result, the concentration of Mn was low in the dredged trench off Makuhari, whereas there was no significant difference in Fe. It is estimated that the oxidation-reduction potential in the dredged trench remained to the extent that Fe precipitation occurred but no Mn precipitation occurred. In addition, the values of Th/U and Ce/U ratios at each site existed in different places on a same line. It is found that the magnitude of the reductive environment increased in the order of flat seafloor off Yokohama < flat seafloor off Makuhari < dredged trench off Makuhari. This trend is in good agreement with water quality data and estimation results by $^{57}$Fe Mössbauer spectroscopy.
The Fukushima nuclear accident released a large amount of radioactive nuclides into the environment. Among them, short half-life radionuclides, for example $^{110m}$Ag, would be paid less attention to; however, those are critical for external and internal exposure just after the accident. In this study we prepare carrier-free radioactive silver, $^{105}$Ag, as tracer nuclide, which is suitable for plant and animal experiment. $^{105}$Ag was produced from cadmium through photo-nuclear reaction. An aliquot of CdCl$_2$ of 2.5 g in a quartz test tube was dehydrated in a muffle furnace and encapsulated under vacuum. The test tube was irradiated with photons generated by the bombardment of platinum sheets with 30 MeV-electrons. $^{105}$Ag was purified by precipitation and anion exchange chromatography. Cadmium was removed by precipitation at pH 11 in chloride solution, while silver remained in the solution as complex ion. Cadmium and silver was once adsorbed onto anion exchange resin, then cadmium was washed out with 0.1 M HCl and finally silver was recovered with 0.1 M HNO$_3$. The resulting product showed that the specific activity of $^{105}$Ag was 6.8 (kBq/mL) and the concentration of cadmium was 10 ppb.
P57 Non-destructive elemental analysis of Ni based super alloys by using time of flight prompt gamma-ray analysis
TOH, Y.1, HUANG, M.1,2, SEGAWA, M.1, MAEDA, M.1, TSUNEYAMA, M.1,3, KIMURA, A.1, NAKAMURA, S.1
(1JAEA, 2IMP, 3NAT.)

The Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) has been developed for measurements of prompt gamma-ray analysis (PGA), nuclear cross-section data and nuclear astrophysics at BL04 of the Material and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). We have developed an analytical technique which combines prompt gamma-ray analysis with the time-of-flight technique by using an intense pulsed neutron beam. We have conducted several experiments of Ni-based super alloys samples, and have analyzed 3 types of spectra PGA, TOF(NRCA) and TOF-PGA. Presentation will describe the detail of the experimental results and new device (neutron filter), the current status of the developments and future prospects.

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P58 LET Dependence of Reactive Oxygen Species Generations in an Aqueous Sample Irradiated by Heavy-Ion Beam
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The amounts of reactive oxygen species generated in aqueous samples by irradiation with X-ray or clinical carbon-ion beams were quantified. Hydroxyl radical (•OH), hydrogen peroxide (H₂O₂), and the total amount of oxidation reactions were measured by electron paramagnetic resonance-based methods. •OH generation was expected to be localized on the track/range of the carbon-ion beam/X-ray, and mM and M levels of •OH generation were observed. Total •OH generation levels were identical at the same dose irrespective of whether X-ray or carbon-ion beam irradiation was used, and were around 1.2–1.3 μmol/L/Gy. However, sparse •OH generation levels decreased with increasing linear energy transfer, and were 0.61, 0.53, and 0.38 μmol/L/Gy for X-ray, 20 keV/μm carbon-ion beam, and 80 keV/μm carbon-ion beam sources, respectively. H₂O₂ generation estimated by TEMPOL decay was as 0.21, 0.57, and 0.35 μmol/L/Gy, for X-ray, 20 keV/μm carbon-ion beam, and 80 keV/μm carbon-ion beam sources, respectively, whereas the H₂O₂ generation estimated by Fenton-reaction-ability was as were 0.22, 0.26 and 0.29 μmol/L/Gy, respectively. TEMPOL decay may only reflect the dense H₂O₂ generation. The amounts of total oxidation reactions were 2.74, 1.17, and 0.66 μmol/L/Gy, respectively. The generation of reactive oxygen species was not uniform at the molecular level.
Kohitsugire are ancient paper sheets containing elegant calligraphy. They were originally pages or parts of ancient manuscripts, pictures or sutras. The old calligraphy written before the 14th century hardly remain as complete books or scrolls; therefore, kohitsugire potentially has high academic value. Most of kohitsugire have traditional certificates which identify the calligraphists. However, recent paleographical study shows that some of them are not genuine handwriting. Therefore, we applied radiocarbon dating to such kohitsugire attributed to famous persons: Ono no Tohu (894-966), Saigyo (1118-1190) and Retired Emperor Gotoba (1180-1239). The radiocarbon dating indicated that all of them were not their genuine handwritings. They were written by different persons in a few century later. It suggested that many copies and counterfeits are in circulation among kohitsugire attributed to famous persons.

In the previous studies, we focused on deposition following valence changes and investigated the electrolytic reduction of U(VI) in weak acid solutions. We found that U(IV) deposits form on the electrode surface by the electrolytic reduction of U(VI) to U(V). Also, we proposed the deposition mechanism that a U(IV) hydroxide formed as an intermediate and transforms to U(IV) oxide as a terminal product. In this study, we investigated the electrolytic deposition of Np(V) by measuring electrochemical quartz crystal microbalance (EQCM) to understand the difference in electrolytic deposition among actinides. The changes of the reduction current and deposition amount with time during -0.75 V was applied to reduce Np(V) were measured. The reaction stopped when the electrode surface was covered with Np deposits. This behavior is different from the deposition of U that the reaction continues even if deposits form on the electrode surface. From the relationship between the quantity of electricity of electrolytic reduction of Np(V) and the weight of deposits in the pH 4 solution, we found that the specie of deposits is highly likely to be Np(IV) dioxide (NpO₂) and the NpO₂ on the electrode surface prevent from reducing Np(V) to Np(IV) because of high electrical resistance of NpO₂.