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KURRI Progress Report 2015

The Research Reactor Institute, Kyoto University

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Preface

This report covers research activities of Kyoto University Research Reactor Institute (KURRI) for the fiscal year 2015, from April 2015 to March, 2016. Researchers under the joint use program of KURRI had difficult time in this period because of the shutdown of KUR and KUCA since 2014. In addition, Tracer Laboratory had major renovations. Nevertheless, researchers made remarkable achievements in the field of research activities with the Electron Linear Accelerator, the Co-60 Gamma-ray Irradiation and other facilities. BNCT research group also continues to work for the development of the Accelerator-based BNCT system. This is the important first step for the future that BNCT treatment will be available in a hospital. They have made good progress with a big mission to shift clinical trial to medical treatment.

In spring, 2016, KURRI obtained approval from the Nuclear Regulation Authority (NRA) to restart KUCA. KUR is the final stage to pass safety assessment at present. After the Fukushima nuclear Accident in 2011, the NRA formulated the New Regulatory Requirements. All the Japanese nuclear facilities have to apply it for the safety review. The NRA also requested research reactors to take strict assessment as same level as nuclear power plants do. At the time of writing this manuscript, we expect both facilities will restart the operation for the research utilization by the end of fall in 2016. I would like to express my heartfelt thanks to joint researchers for their strong patients and to our staffs and all who contributed their great efforts for gaining the approval.

The long shutdown of all research reactors in Japan had a large influence on nuclear/neutron research and education. Especially, cancer patients suffered, because they could not receive medical treatments with the reactor-based BNCT. In the point of human resource development, universities did not provide students practical nuclear education programs using nuclear facilities before their graduation. The essential problems of those educational circumstances were pointed out by many experts.

The biggest mission for us is the restart of KUR and KUCA at present. We recognized that we faced serious problem to satisfy the social needs and demands in the view of safe and security. It is apparent that these issues impose a substantial burden on KURRI which is just one of institutes in one university. We have to seek for ways to keep capable people in order to achieve safety management and to reinforce our nuclear security.

In the end, we would like inform a new pledge in the latest Nuclear Security Summit, held in Washington, organized by President Barack Obama. The Japanese government agreed with U.S. to remove and transfer high enriched uranium (HEU) fuels from KUCA to the United States. In the near future, HEL will be replaced with low enriched uranium (LEU) fuels. It would be tough burdens for us, but we are honored that the importance role of KUCA as a research and education facility was confirmed in the top level agreement of between Japan and U.S. governments.

Kumatori, July 14, 2016 Yuji Kawabata Director, KURRI

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ANNUAL SUMMARY OF EXPERIMENTAL RESEARCH ACTIVITIES

I-1. PROJECT RESEARCHES

Irradiation Effects on Nuclear Advaced Materials Irradiated by Particles with High Energy

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OBJECTIVES: High dose neutron irradiation is usually used to investigate radiation effects in solid materials and develop new materials. Unfortunately, KUR could not supply the neutron irradiation field and the laboratory for neutron-irradiated samples was closed in 2015. The research activities were severely restricted. Most of researchers had to change their research purpose to get the results. Therefore, instead of neutron irradiation, electron and γ ray irradiations were used to estimate the damage induced by the irradiation in materials.

RESULTS: The carried out research subject (ARS) and the name of co-researches in each ARS are listed below. ARS-1

Modification of the yellow luminescence in gamma-ray irradiated GaN bulk single crystal

(K. Kuriyama, A. Shikawa, N. Nishikata, K. Kushida, and Q. Xu)

The results show that gamma-ray induced shallow donor relating to nitrogen vacancy is located at about 50 meV below the conduction band.

ARS-2

Photoluminescence of natural and synthesized calcites (T. Awata, S. Tanaka and Q. Xu)

Photoluminescence (PL) of natural calcites and impurities of these, and compared spectra of artificially synthesized calcite in chemical reaction have been measured.

ARS-3

Thermal diffusivity measurement of small specimens to achieve the measurement of heavily neutron irradiated tungsten

(M. Akiyoshi and Q. Xu)

The measurement results of thermal diffusivity between small specimen and 10 mm standard specimen are different. The result of small specimen is small even above 373 K compared with 10 mm standard specimen.

ARS-4

Electron irradiation induced damage struttre in

intermetallic alloys

(F. Hori, Y. Ueno, T. Ishiyama, K. Kobayashi, A. Iwase, K. Ohsawa, Q. Xu and N. Abe)

Before irradiation, the single Al-vacancy remains in Fe-50Al alloy. After irradiation, however, the differnent types of vacancy were produced into Fe-Al compound alloy.

ARS-8

Effects of high energy particle irradiation on hydrogen retention in refractory metals

(K. Tokunaga, M. Matsuyama, S. Abe, H. Osaki, K. Araki, M. Hasegawa, K. Nakamura, Q. Xu and K. Sato) The amount of tritium increases with increasing the irradiation dose. In addition, tritium is easy to diffuse parallel direction for rolling direction after irradiation.

ARS-9

Positron annihilation spectroscopy of Ferritic/Martensitic steels irradiated with protons and spallation neutrons (K. Sato, Q. Xu, T. Yoshiie, Y. Dai and K. Kikuchi) Vacnacy clusters are formed by the irradiation in F82H. The vacancy clusters including small and latge amount of He atoms up to 15 dpa of irradiation.

ARS-10

Positron annihilation behavior in Fe-Cr alloys

(R. Kasada, K. Sato and Q. Xu)

In unaged $Fe_{0.91}Cr_{0.09}$ and $Fe_{0.84}Cr_{0.16}$ alloys, there are not enhancement of positron density. In contrast, an enhancement of annihilation in Fe cell in other Fe-Cr alloys, such as, $Fe_{0.7}Cr_{0.3}$, $Fe_{0.55}Cr_{0.45}$ and $Fe_{0.5}Cr_{0.5}$.

CONCLUSIONS: The defects induced by irradiation degrade the physical mechanical properties of solid materials. Some experimental results are important for development of nuclear industry. For example, the results of ARS-8 indicate that the retention of tritium, which is the isotope of hydrogen, in refractory metals used in fusion reactor may be an important issue because the plasma-facing materials suffer two types of damage: displacement damage caused by high energy neutrons and surface damage.

PR1-1 Modification of the Yellow Luminescence in Gamma-ray Irradiated GaN Bulk Single Crystal

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INTRODUCTION: Examining the defects caused by various radiations to GaN under the space environment is important. In our previous study [1, 2], we reported that the energy levels relating to nitrogen vacancy (V_N) and gallium vacancy (V_{Ga}) were induced by neutron and proton irradiated GaN. The neutron irradiation has been used as the neutron transmutation doping into semiconductors such as GaAs [3], GaP [4], and GaN [5]. Atoms in semiconductors mainly transmute by a (n, γ) reaction. Therefore, to survey the radiation effect of gamma ray alone is meaningful. In the present study, we report the variation of the yellow luminescence (YL) in GaN bulk single crystals by gamma-ray irradiation.

EXPERIMENTS: GaN bulk single crystals with a thickness of 450 \pm 50 μ m were purchased from Furukawa Co. Ltd. The crystals were irradiated at room temperature with gamma-rays of 1.17 and 1.33 MeV from a cobalt-60 source of Kyoto University Research Reactor Institute. Samples were irradiated with an absorption dose rate of 1.771 KGy/h. Total gamma-ray dose was 160 kGy. The resistivity varied from 30 Ω cm for an un-irradiated sample to 10⁴ Ω cm for gamma-ray irradiated one. Photoluminescence (PL) spectra were measured at 77 K using a He-Cd laser.

RESULTS: The band edge emission was observed at 3.48 eV in both un-irradiated and gamma-ray irradiated samples. The YL from the un-irradiated GaN with a peak at 557 nm (2.22 eV) was observed at around 440 nm to 800 nm, whereas that of the gamma-ray irradiated GaN showed a peak at 532 nm (2.33 eV) although the YL spectrum is almost overlapped with un-irradiated ones. Therefore, the modification of YL observed here would be attributed to a transition from the shallow donor induced by the gamma-ray irradiation to a native V_{Ga} . Since the usual YL has been proposed as a transition from a shallow donor to V_{Ga} lo-

cated at about 1.1 eV above the valence band [6], the gamma-ray induced shallow donor is located at about 50 meV bellow the conduction band. This energy level is close to a donor level ($64 \pm 10 \text{ meV}$ [7]) relating to $V_{\rm N}$. Figure 1 shows the YL model estimated from the photoluminescence at 77 K.



Fig.1 Yellow luminescence estimated from the photoluminescence at 77 K. The gamma-ray induced shallow donor relating to nitrogen vacancy is located at about 50 meV below the conduction band.

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Photoluminescence of Natural and Synthesized Calcites

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INTRODUCTION: Thermoluminescence (TL) of natural calcite irradiated by gamma rays have only one orange emission peak and that may be originated from an impurity of Mn^{2+} in calcite [1-2]. At photoluminescence (PL) of natural calcite irradiated UV rays, the spectra of PL have 620nm emission peak and 270nm emission peak which is related Mn^{2+} and Pb^{2+} , respectively [3]. In the case of calcites, it has been shown the impurity plays a major role for luminescence. In this study, we first have measured PL of natural calcites and impurities of these, and compared spectra of artificially synthesized calcite in chemical reaction with resulted major impurities.

EXPERIMENTS: Natural calcite with colored pink was collected at Colorado State in USA. Artificial calcite was synthesized by a chemical reaction method in water solution [4]. 1.5% of (NH₄)₂CO₃·2H₂O(1.5g in 100mL pure water) and 1.8% of CaCl₂·6H₂O (2g in 100mL pure water) with 0.01g element impurity were mixed at room temperature. After mixed, these solutions were precipitated by holding for two weeks and filtering. After filtering, samples were shaped by a tablet machine. PL spectra have been taken using PL-84 (Seishin, SOEX1702/04, PM:Hamamatsu R928) with He-Cd laser (Kinmon 325nm). ICP-AES (iCAP6300Duo, Themo Fisher) was also performed to measure impurity elements concentration of calcite.

RESULTS and DISCUSSION: Figure 1 shows the PL spectra of Colorado natural calcite irradiated 325nm UV rays. It is shown that a spectrum has three peaks at 396nm, 564nm, 760nm and 564nm peaks. 564nm peak is





a strongest and most sharp (FWHM=4%). From ICP-AES results, Colorado calcite has impurities (ppm) of Sr (5.1), Mn (N.D.) and Pb (0.05). Nevertheless almost calcite has large amount of Mn impurity, Colorado calcite has very little amount. There is no peak at 620nm in figure 1, that is related to little Mn amount. Photoluminescence data reference [5] reported that emission peak at 560nm for defect center of carbonate ion. From experimental results, we have synthesized calcite artificially with controlled impurities. Fig 2 shows that PL spectra of synthesized calcite with an impurity of Sr and Sr + Mn. It is shown that both



Fig. 2 Photoluminescence spectra of synthesized calcite with an impurity of Sr and Sr + Mn.

spectra have four peaks at 427nm, 493nm, 560nm and 756nm. Comparing PL spectra of natural and synthesized calcite, both spectra are almost same. PL spectra of synthesized calcite with Sr and Sr + Mn are almost same and there is no effect of presented Mn ion. Sidike et al., reported [3] that Mn ion cannot be an activator alone for PL, but can be activator with co-activator as Ce ion or Pb ion which can absorb UV rays. Relatively large amount impurity in Colorado calcite, Sr ion, is not activator but also co-activator for Mn ion.

ACKNOWLEDGEMENT: We would like to thank Professor S. Fukutani (KURRI) for using ICP-AES on this study.

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PR1-3 Thermal Diffusivity Measurement of Small Specimens to Achieve the Measurement of Heavily Neutron Irradiated Tungsten

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INTRODUCTION:

Usually, heat is mainly carried by free electrons in metals. On the other hand, it is well known that irradiation induced damages in ceramics showed significant degradation in thermal diffusivity unlike metals, because in ceramics, heat is mainly carried by phonon that is scattered by irradiation induced defects. In these days, development of materials for divertor in future fission reactor is very important and concerned study. Most of studies are focused on SiC ceramics and tungsten based materials. Tungsten is a metal, of cause, however heat is carried by phonon in tungsten partly. Then, thermal diffusivity of tungsten after severe neutron irradiation must be estimated in the same way as ceramics.

In addition, tungsten shows far stronger radioactivity after a neutron irradiation than SiC ceramics. In the PHENIX project for development of divertor materials in fusion reactor, many SiC and W specimens were loaded to HFIR reactor in ORNL, USA. To reduce the level of radioactivity, specimens for thermal diffusivity measurement is limited to 6mm in diameter or further small 3mm in diameter disk specimens with 0.5mm thickness.

Measurement of thermal diffusivity after the neutron irradiation in HFIR is must to be operated using Netzsch LFA-457 Micro Flash thermal analyzer in ORNL, which can measure 6mm disk specimens using commercial jigs, while measurement of 3mm small disk is not certified.

EXPERIMENTS:

To achieve a measurement of small specimens 3mm in diameter using Netzsch LFA-457, special jigs were designed and manufactured. The jigs shade the incident laser flash to 2.4mm in diameter and also cut the infrared respond signal radiated from the back side of the specimen to 2.6mm in diameter.

Measurements of thermal diffusivity using small disks were performed at ORNL, USA using Netzsch LFA-457, and also 10mm in diameter standard sample was measured using Ulvac TC-7000 at the Radiation Laboratory, Uji, Kyoto University. Specimens were ITER grade pure tungsten (supplied by A.L.M.T. Corp.) with the size of 6mm and 10mm in diameter and commercial grade pure tungsten (supplied by Nilaco) with the size of 3mm and 10mm in diameter. The measurements were operated at room temperature to 773K using Netzsch LFA-457, and to 973K using Ulvac TC-7000, respectively.

RESULTS:

Fig.1 shows thermal diffusivity of ITER Grade tungsten with the size of 6mm and 10mm in diameter. The measurement result of a 6mm sample showed large dispersion at around room temperature. The laser flash method uses uprising curve of temperature on the back side surface after a laser flashing, and the Netzsch LFA-457 system uses IR detector to measure the temperature. Intensity of the IR is changed with a temperature according to the Planck radiation formula, and also with measuring area of a specimen. The back side area of a 6mm specimen is only 36% of a 10mm specimen. Then the wave-shape from the IR detector was noisy at around room temperature, while above 373K, the intensity of IR was increased enough to evaluate the thermal diffusivity correctly.

This result is almost same for a 3mm specimen shown in Fig.2, however measurement result of this small specimen is obviously small even above 373K compared with 10mm standard specimen. The difference in the shape of these specimens is not only the surface area but also the thickness, 0.973mm for 10mm diameter and 0.598mm for 3mm diameter specimen. The later thickness is too thin to evaluate correctly for tungsten before neutron irradiation. The difference between the thermal diffusivity of 10mm diameter and 3mm diameter getting smaller at higher temperature, because the uprising time of IR after the laser flashing getting longer and analyzed better. Neutron irradiated tungsten shows smaller thermal diffusivity than the unirradiated specimens here, so the difference is expected to be enough small with the specimen of 0.5mm thickness.







Fig.2 Validation of thermal diffusivity measurement using a specimen of 3mm in diameter.

PR1-4 Electron Irradiation Induced Damage Structure in Intermetallic Alloys

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INTRODUCTION: It is well known that intermetallic compound alloys possibly include more than two types of vacancies basically, that is A-vacancy and B-vacancy in A-B compound alloy. Thermally equilibrium defects and chemically deviated structural defects in these compound alloys are very complicated. However, the amount of defect and the defect structure affects the various characteristic features, such as strength and electronic conductivity and so on. Moreover, first principle calculation shows that different number of hydrogen atoms can be trapped by Al- or Fe-vacancy in B2 type Fe-Al alloy. On the other hand, although the electron irradiation produces vacancies and interstitials in metallic alloys, the threshold energy of displacement for each elemental atom in compound alloys is not necessarily same. Then the defect type introduced by electron irradiation depends on its incident energy. Kosugi et al. reported that the type of defects produced by electron irradiation in B2 type Fe-Rh intemetallic alloy changes with increasing of incident electron energy [1]. Then we supposed that radiation induced vacancy type defects can be controlled by changing the energy of electron irradiation. In this study, we have performed electron irradiation with different energy for Fe-Al alloys.

EXPERIMENTS: Fe-50%Al alloy specimens with B2 structure were prepared by arc melting method. Sliced samples with the thickness of 0.5 mm were annealed at 973 K for 120 h followed by air-quenched in vacuum. These specimens were irradiated with 2 MeV electron to the fluence of 1×10^{17} and 1×10^{18} /cm² at JAEA-Takasaki and with 9 MeV electron to the fluence of 5×10^{17} and 3×10^{18} /cm² at KURRI, Kyoto University. In both cases, irradiations were carried out at about 330 K controlled by water flow system. After and before irradiation, samples were examined by positron annihilation lifetime and coincidence Doppler

broadening measurements. The positron lifetime spectra were analyzed by using POSITRONFIT program. Also, the positron lifetimes in B2 type Fe-Al alloys were calculated using the superimposed atom method [2].

RESULTS: Before irradiation, positron lifetime of this alloy was 186 psec showing that single vacancy still remains in this alloy. This positron lifetime value is close to that in Al-vacancy by the calculation. The positron annihilation lifetime value of this alloy increases to 200 psec after 2 MeV electron irradiation and decreases to 172 psec after 9 MeV electron irradiation, respectively. Also, the Doppler broadening S-parameter showed different changing trend for each electron irradiation with different energy. That is S-parameter increases for 2 MeV irradiation and decreases for 9 MeV irradiation case. Figure 1 shows the changing relation between positron lifetime and S-parameter for each energetic electron irradiations. This result suggests that different types of vacancy were produced into Fe-Al compound alloy by electron irradiation with different energies.



Fig. 1 Positron lifetime and S-parameter relation for before and after electron irradiated Fe-Al alloy.

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Effects of High Energy Particle Irradiation on Hydrogen Retention in Refractory Metals

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INTRODUCTION: It is of importance to clarify phenomena of implantation, retention, diffusion and permeation of tritium on surface of the armor materials of the first wall/blanket and the divertor on fusion device from a viewpoint of precise control of fuel particles, reduction of tritium inventory and safe waste management of materials contaminated with tritium (T). Refractory metals such as tungsten (W) is potential candidate for an armor of the first wall and the divertor plate of the fusion reactor because of its low erosion yield and good thermal properties. The armor material will be subjected to heavy thermal loads in the steady state or transient mode combined with high energy neutron irradiation that will cause serious material degradation. In addition, high energy runaway electrons would bombard the armor materials along the equatorial plane in fusion device. It is considered that these cause radiation damage and enhance tritium retention. In the present works, T exposure experiments have been carried out on W samples which were irradiated by high energy electrons using LINAC in KURRI of Research Reactor Institute, Kyoto University to investigate effects of high energy electrons irradiation and microstructure on tritium retention of W. In the repot of the last fiscal year, results of T exposure experiments on ITER grade W which was fabricated by powder metallurgy. In the present report, results of T exposure experiments on W which was fabricated by Hot Isostatic Pressing (HIP-W) and comparison of results with ITER grade W and HIP-W will be described.

EXPERIMENTS: W samples used were ITER grade W (IG-W) and Hot Isostatic Pressing W (HIP-W). In the case of IG-W, one was W sample (ITER grade W(1)) which the surface were manufactured to be oriented parallel to the rolling surface and rolling direction. The other W sample (ITER grade W(3)) which the surface were manufactured to be oriented perpendicular to the rolling surface and rolling direction. On the other hand, structure of HIP-W is equiaxed deriving from the nature of the consolidation process. The sizes of W samples were 10mm x 10mm x 1mm. The surface of the both samples were polished to be mirrored. High energy electrons irradiation has been carried out using LINAC in KURRI of Research Reactor Institute, Kyoto University. An energy of electron irradiated was 10 MeV and DPA was

3.26 x 10-3. Temperature during the irradiation was measured by thermocouples which was contacted with a backside of the W samples. After the electron beam irradiation, T exposure experiments have been carried out using a T exposure device in University of Toyama. Pressure of the T gas was 1.3 kPa and T exposure was kept for 4 h. T concentration in the gas was about 5 %. Temperatures of pre-heating and T exposures were 100 oC. After the exposure to T gas, T amount retained in surface layers of the sample was evaluated by β -ray-induced X-ray spectrometry (BIXS) and imaging plate (IP) measurements.

RESULTS: In the condition of 10 MeV electrons irradiation, electrons go through in 10 mm thickness W sample. As a result, uniform defects were formed in the W samples. Fig. 1 shows IP images of HIP-W, IG-W(3) and IG-W(1). It can be seen that amount of T near surface depends on kinds of W. Fig. 2 shows amount of T as a function of DPA. In general, the amount T increases with increasing DPA. In addition, it is considered that T is easy to diffuse parallel direction for rolling direction from comparing with IG-W(3) and IG-W(1) after the electron irradiation.



Fig. 1. Tritium image of HIP-W, ITER grade W(3) (IG W(3) and ITER grade W(1)(IG W(1)). Upper parts are standard samples.



Fig. 2. Amount of tritium as a function of DPA

Positron Annihilation Spectroscopy of Ferritic/Martensitic Steels Irradiated with Protons and Spallation Neutrons

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INTRODUCTION: In fusion reactors and spallation neutron sources, irradiation damage is significantly more serious than fission reactors. There is a need for use of irradiation resistant materials in high neutron dose facilities. Due to its good thermal and mechanical properties, the reduced-activation ferritic/martensitic steel, F82H is a candidate material for the first wall and blanket of the future fusion reactor [1]. There have been a number of studies on the void swelling behavior of structural materials after neutron irradiation [2,3]. Recent theoretical and experimental analyses have revealed the importance of incubation period, a transient stage before the steady growth of voids. Positrons are very sensitive to vacancy type defects, and even single vacancies can be detected in positron annihilation lifetime measurements. In this study, defect structures in both F82H and SS316 steels during the incubation period were studied after proton irradiation using positron annihilation lifetime measurements.

EXPERIMENTS: The reduced activated ferritic/martensitic steel F82H was used in this experiment. F82H was irradiated in the second program (STIP-II). More details of the STIP-I and STIP-II experiment were previously reported [4]. Table 1 shows sample ID, irradiation dose, average irradiation temperature and calculated production of H and He. It should be noted that the actual H contents can be much lower than the calculated values due to fast diffusion of H in steels [5]. The chucks of tensile test samples of F82H were used for positron annihilation lifetime measurements at Paul Scherrer Institut.

RESULTS: Before irradiation, the long lifetime of about 150 ps denotes the lifetime of the dislocations and the lath boundaries. Vacancy clusters are formed by the irradiation. In F82H, He bubbles formed by the irradiation above 443 K and about 500 appm He are visible by TEM. He bubble size increases and the concentration decreases with increasing the irradiation dose and temperature by TEM. However, the long lifetime intensity does not increase with increasing the irradiation dose. Therefore, it is also possible to detect invisible He bubbles by positron annihilation lifetime measurements. The positron annihilation lifetime of vacancy clusters de-

creases to less than 200 ps when the vacancy clusters contain a large amount of He atoms. The short and long lifetime at 5.9 dpa is almost the same as the calculated lifetime of V1 and V15, respectively, due to a lot of gas atoms are produced during the irradiation. Therefore, the long lifetime and short lifetime up to 15 dpa denote vacancy clusters including small and large amount of He atoms, respectively. Decrease of long lifetime and mean lifetime up to 15 dpa is due to the absorption of He atoms. The concentration of vacancy clusters including small amount of He atoms decreases up to 10 dpa because they absorb He atoms and are classified into the short lifetime. After that, the concentration of vacancy clusters including small amount of He atoms increases again. In this stage, small vacancy clusters, which are classified into the short lifetime up to 10 dpa, grow more prominently and are classified into the long lifetime. From TEM observation, He bubbles grow with increasing irradiation dose above 10 dpa, but the positron lifetime does not increase. The absorption of He atoms is also large. If V₃₄₁ (about 1.5 nm in diameter) contains 341 and 682 He atoms, the positron lifetime changes from 441 ps to 242 and 187 ps in Fe, respectively [6]. These results suggest that vacancy clusters contains more He atoms than number of vacancies which make up the vacancy clusters

Table 1. Irradiation condition of F82H.

ID	Tempera- ture (K)	Irradiation dose (dpa)	He (appm)	H (appm)
L11-L L12-L	371	5.9	413	1680
L11-Н L12-Н	397	8.5	650	2530
L13-L L14-L	406	9.2	715	2850
L13-Н L14-Н	448	12.3	1015	4200
L15-H L16-H	513	17.2	1505	6200
L17-H L18-H	560	20.4	1795	7720

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above 15 dpa.

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INTRODUCTION: Ferritic steels containing Cr are expected to be used for the first-wall component of the fusion reactors as well as for the fuel pin cladding of the Generation IV nuclear fission systems [1]. However, high-Cr steels may suffer from thermal aging embrittlement, which is well-known 475 °C embrittlement. This is mainly due to hardening phenomenon through the phase separation of Fe and Cr as shown in the phase diagram. In the previous study [2], we applied a positron annihilation spectrometry to detect the phase separation in the Fe-Cr alloys after thermal aging at 475 °C. Currently, this collaborative research is investigating neutron irradiation effect on the positron annihilation behavior of Fe-Cr alloys. Here, we reports a new model to explain the positron annihilation behavior in Fe-Cr binary alloys.

Experimental Procedure: Positron annihilation coincidence Doppler broadening (CDB) measurement were performed at the hot laboratory of KURRI.

Results and Discussions: Based on the theoretical model of positron annihilation in binary alloys suggested by Lock and West [3], we modeled the effect of Cr concentration and aging on positron annihilation in disordered (unaged) Fe-Cr alloys. The expression for the W-parameter of the Fe-Cr alloys may be represented by

$$W_{FeCr} = f_{Fe}^{+} W_{Fe} + f_{Cr}^{+} W_{Cr}$$
(1)

as the weighted sum of the W parameter of the constituent pure metals (W_{Fe} and W_{Cr}). The fractions *f* denote the fractions of positron density in the Fe cell and Cr cell. The normalized W-parameters of Fe_{1-x}Cr_x alloys can be written as following equation:

$$\frac{W_{FeCr} - W_{Fe}}{W_{Cr} - W_{Fe}} = f_{Cr}^{+} = 1 - f_{Fe}^{+} = 1 - (1 - x)\eta_{Fe} \qquad (2),$$

where η_{Fe} is the enhancement of positron density in the Fe cell of Fe-Cr alloy compared with the constituent pure Fe.

Figure 1 presents the normalized W-parameter of the CDB measurement against the Cr content *x* in the Fe_{1-x}Cr_x alloys. In Figure 1 the curve 1, which linearly connects the Fe and Cr, represents the simplest case that corresponds to no enhancement of positron density in the Fe-Cr alloys. The unaged Fe_{0.91}Cr_{0.09} and Fe_{0.84}Cr_{0.16} are well fitted to the curve 1. In contrast, the other unaged Fe-Cr alloys indicate an enhancement of annihilation in Fe cell. The nonlinear function of Cr content for the unaged Fe_{1-x}Cr_x alloys suggests a presence of preferential positron annihilation (PPA) in Fe cell at their disordered state. Further experiments and discussion on the aged Fe-Cr alloys are now going.



Fig. 1 Normalized W parameter of Fe-Cr alloys.

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Development on Neutron Imaging Application

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1. Objectives and Allotted Research Subjects

Neutron imaging provides valuable information which cannot be obtained from an optical or X-ray imaging. The purpose of this project is to develop the imaging method itself and also the experimental environment for expanding the application area of the neutron imaging. The allotted research subjects are as follows:

- ARS-1 Measurements of Multiphase Dynamics by Neutron Radiography (Y. Saito *et al.*)
- ARS-2 Visualization and Measurement of Flow Behavior in Industrial Equipment (N. Takenaka *et al.*)
- ARS-3 Visualization and Measurement of Adsorption/Desorption Process of Ethanol in Activated Carbon Adsorber for Adsorption Heat Pump (N. Asano *et al.*)
- ARS-4 Neutron Radiography on Tubular Flow Reactor for Supercritical Hydrothermal Synthesis of Nanoparticles (T. Tsukada *et al.*)
- ARS-5 Characteristics of the Void Fraction under Transient Condition (H. Umekawa *et al.*)
- ARS-6 Estimation of the Frosting and Defrosting Phenomena by Using Netron Radiography (R. Matsumoto *et al.*)
- ARS-7 Neutron imaging and optics development using simulation of VCAD Systems (Y. Yamagata *et al.*)
- ARS-8 Water and Salt Distribution in a Rice Hull Medium under Sodium Chloride Solution Culture (U. Matsushima *et al.*)
- ARS-9 Measurement of Water Content in Hardened Cement Paste by Neutron Imaging (T. Numao *et al.*)
- ARS-10 Hydrazine Thickness Measurement by Neutron Radiography at a Catalyst Bed during Operation (H. Kagawa *et al.*)
- ARS-11 In-situ Neutron Radiography Investigation on the Hydraulic Behavior of High Strength Cement Paste under High Temperature (M. Kanematsu *et al.*)
- ARS-12 Evaluation of coolant distribution in a flat heat-pipe type heat spreader (K. Mizuta *et al.*)
- ARS-13 Visualization of Organic Materials for Development of Industrial Applications (A. Uritani *et al.*)
- ARS-14 Visualization of Coolant Flow in a Micro-Structured Wick (Y. Tsuji *et al.*)

2. Main results and the contents of this report

Due to the termination of KUR, no neutron imaging

has been performed for fiscal 2015. However, some results have been obtained by analyzing existing experimental results or by performing X-ray imaging.

ARS-2 applied X-ray imaging to two-phase flow across horizontal tube bundles of in-line and staggered. X-ray radiography was employed for the measurement of void fraction distributions. The transmitted X-ray was converted to visible rays using an image intensifier, and the 10-bit gray-scale radiographs were taken using a high-speed camera (MotionPro Y-4 Lite, IDT Inc.) with an array of 1024×1024 pixels and with a pixel size of 170 µm. The flame rate was set at 30 fps and 450 radiographs which correspond to 15 sec were taken in each condition. The X-ray was generated at 80 kV and 5 mA.

heat transfer study on an adsorption refrigerator. In this ARS, visualization of adsorption amount distributions of ethanol in an activated carbon powder bed. Experimental results show that transient behaviors of adsorption amount distribution could be clearly observed by neutron radiography.

ARS-3 visualized transient change of adsorbed refrigerant amount during an adsorption process by using neutron imaging. From the measurement results effect of the metallic fins was investigated for improvement of heat diffusion in adsorption process.

In ARS-4, flow behavior of heavy oils in a packed-bed reactor was investigated by using B-4 neutron imaging facility. In this work, it was revealed that the visualization of flow behaviors of heavy oils in the packed-bed reactor at high temperature and high pressure is feasible by neutron radiography.

ARS-5 conducted frosting and defrosting experiments by using neutron/X-ray imaging. On the cooled surface of a heat exchanger, the frost grows with a spatial distribution that is determined by the temperature and humidity profiles of the air and the flow pattern in the heat exchanger. This means that the mass transfer coefficient for frost formation has both temporal and spatial distribution. However, in conventional methods of measuring the frost accumulation rate, the frost is scraped from the surface, or the frost accumulation is estimated from the difference between the absolute humidity before and after the heat exchanger. In the present study, the frost formation and defrosting process was estimated by the two ways by neutron and X-ray radiographies at KUR B-4 radiation port. **PR2-1**

Void Fraction Distritbution around a Tube in Two-phase Flows Across Horizontal Tube Bundles

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INTRODUCTION: Heat exchangers such like shell and tube type are commonly used in process industry, chemical industry, and geothermal power plant. The void fraction in the flow channel are important parameters for predicting the flow characteristics, fluid oscillation and heat transfer. To clarify the two-phase flow structures, many experimental studies have been carried out in a rectangular channel with the tube bundle [1-4]. However, many of these investigation evaluate local void fraction at several measurement positions in the tube bundles, and the two-dimensional void fraction distributions in the bundles has not been sufficiently carried out.

In this study, two-dimensional void fractions were measured by using X-ray radiography in two-phase flow across horizontal tube bundles of in-line and staggered. Difference of void fraction distribution in each flow regime and bundle arraignment were compared.

EXPERIMENTS: The test section was vertical duct with a cross-section of $90 \times 90 \text{ mm}^2$. Air was injected into the test section through two porous tubes located at the bottom of the test section. Two different tube bundles were employed for the measurements. One was an in-line tube bundle with 8 rows of three full tubes and two half tubes. And the other was a staggered tube bundle with 8 rows. The tube in both bundles were 90 mm long and the outer diameter, *d*, was 15 mm. The tubes pitch, *p*, was 22.5 mm and the pitch-to-diameter ratio, p/d, was 1.5. The experiments were performed at 20–25 °C.

X-ray radiography was employed for the measurement of void fraction distributions. The transmitted X-ray was converted to visible rays using an image intensifier, and the 10-bit gray-scale radiographs were taken using a high-speed camera (MotionPro Y-4 Lite, IDT Inc.) with an array of 1024×1024 pixels and with a pixel size of $170 \ \mu\text{m}$. The flame rate was set at 30 fps and 450 radiographs which correspond to 15 sec were taken in each condition. The X-ray was generated at 80 kV and 5 mA.

RESULTS: Results of two-dimensional void fraction distributions are shown in Fig. 1. The mass flux, *G*, is 200 kg/m²s, and the superficial gas velocity, J_G , is 0.36 and 0.86 m/s. The measurement area is around center tube at 4th row from the inlet of the bundles.

For in-line tube bundle, many bubbles pass travel along the vertical tube gaps, and the void fraction became higher than that at the other region in bubbly flow. Some bubbles passing through the bottom of tubes. As a result, void fraction around the upstream of each tube becomes higher than that around downstream of the tubes. In intermittent flow regime, large bubble intermittently pass through the tubes. As a result, void fraction around the maximum gap and vertical minimum gap increased. The maximum void fraction around maximum and vertical minimum gaps were almost the same. However, void fraction around the horizontal minimum gap is slightly lower than that around maximum and vertical minimum gaps.

For staggered tube bundle, the void fraction around upstream of tubes is slightly high in comparison with the horizontal tube side. Tendency of the void fraction distribution is almost the same with that in in-line tube bundle, however, the void fraction around the bottom of tubes are slightly higher than that in the other region. At intermittent flow, void fraction around bottom of tubes became much higher than the other region. Therefore, it can be confirmed that large bubbles tend to be stagnant around the bottom of tubes. As a result, void fraction has large distribution in comparison with that in in-line tube bundle. Furthermore, the maximum void fraction in staggered tube bundle was higher than that in in-line tube bundle.



Intermittent flow ($J_G = 0.86$ m/s) Fig. 1. Two-dimensional void-fraction distributions in in-line and staggered tube bundles.

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PR2-2

Quantitative Measurement of Adsorbed Ethanol Amount in Activated Carbon Adsorber for Adsorption Heat Pump

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INTRODUCTION: Adsorption refrigerator is one of the efficient tools for waste heat utilization at low temperature. In the refrigerating cycle, adsorption and desorption processes are alternately repeated in batch operation. Since heat removal of latent heat of adsorbed refrigerant is necessary during the adsorption process, improvement of heat diffusion in adsorption layer. Although metallic fins are usually used for the enhancement in the heat diffusion, the amount of inserted fins should be minimized from the standpoint of the downhold of the heat capacity. It is important for the improvement of adsorber to clarify the distribution of adsorbed refrigerant amount in an adsorbent bed during transient conditions of adsorption/desorption. Neutron radiography was applied to visualize the adsorption amount distribution of ethanol in an activated carbon particle bed, and the effect of aluminum fins were evaluated from the measured results.

EXPERIMENTS: Activated carbon and ethanol was used as the adsorbent and refrigerant, respectively. The activated carbon MAXSORB® III produced by Kansai Coke and Chemical Co., Ltd. was used as the adsorbent. Activated carbon was packed in an aluminum frame shown in Figure 1. The container was connected to an ethanol reservoir. The vapor pressure in the container could be maintained by the temperature of the reservoir. The front and back wall was made of thin stainless steel plates to avoid heat diffusion through the walls. The adsorber could be cooled in adsorption process or heated in desorption process by water through a channel in the bottom wall. Two aluminum fins with the thickness of 1 mm were inserted in the adsorbent bed to enhance the heat diffusion. The mass attenuation coefficient of ethanol against thermal neutron was measured as 3.86 g/cm2 in our preliminary experiment.

RESULTS: Transient change of adsorbed refrigerant amount during an adsorption process was measured quantitatively by the umbra method using a checked-type neutron absorber. Figure 2 (a) shows a visualized image with the neutron absorber grid. The inside wall including the fins are indicated by white lines. The measured adsorbed amounts in the center section and temperature in the adsorption bed are shown in Fig. 2 (b) and (c), respectively. At the initial condition, the adsorbent was dry and vacuated. Then, by opening a valve between the ethanol reservoir and the test section, vapor ethanol was supplied to the test section. The horizontal axis of Fig. 2 (a) and (b) shows the elapsed time from the opening of the valve. Just after the start, the adsorbent temperature rapidly increased, and reached around 70 °C due to the strong adsorption. Then, adsorbed refrigerant amount gradually increased with a gradual decrease in the adsorbent temperature due to heat diffusion. The difference in the temperature of the adsorbent between T_{ad2} and T_{ad3} was little. The adsorption amounts in the area of 5, 7, and 8 near the heat transfer surfaces were larger than the amounts in the area of 6 during the transient condition. The fact means that the phenomenon is dominated by heat conduction in the adsorbent bed.



PR2-3 Neutron Radiography on Flow Behavior of Heavy Oils in Packed-Bed Reactor

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INTRODUCTION: With the increase of the demand for petrochemical feedstock and middle distillate, it is desired to utilize heavy crude oils and residues as new resources. However, as heavy oils have high viscosity and poor quality, the upgrading and refining processes of heavy oils are indispensable. For instance, a trickle-bed reactor is commonly used as the residue hydrodesulfurization unit of heavy oils, and thus, it is important to understand the flow behavior of heavy oils through a packed bed of catalyst pellets at high temperature and high pressure for design and optimization of the process. However, because of the reactor which is made of metal and the high-temperature and high-pressure processing, it is impossible to visualize directly the flow behavior inside the reactor. Therefore, we are going to use neutron radiography to visualize the flow behavior of heavy oils in a trickle-bed reactor. In this work, as a first step of such visualization, the flow behavior of bitumen through a packed bed were observed at atmospheric pressure using neutron radiography.

EXPERIMENTS: Fig.1 shows the schematic diagram of a packed-bed reactor, which was made of a SUS316 tube with outer diameter of 1/2 inch and contained alumina particles with 1 or 3 mm diameter. Heavy oil was fed to the reactor through SUS316 1/16 inch tube by using a high-pressure pump. The fourth of five fractions fractionated by vacuum distillation of bitumen was used as a sample. Here, the viscosity of the sample was adjusted by varying the temperature between 40 and $80^{\circ}C$.

Neutron radiography of the packed-bed reactor, i.e., the flow behavior of the sample through the packed bed, was carried out, where the neutron radiographs were taken by an image intensifier and a high speed camera. A thermal neutron beam emitted from the B4 port in KUR was used. The imaging method of neutron radiography and subsequent analysis of the images were almost similar to those in our previous work [1].

RESULTS: Figs. 2 (a) to (c) show the neutron radiographs of flow behaviors of packed-bed reactor, where (a) the temperature T = 69.8°C, the flow rate Q = 0.6 mL/min, and the diameter of particles D = 1 mm, (b) T = 73.0°C, Q = 0.6 mL/min, D = 3 mm, and (c) T = 44.2°C, Q = 0.6 mL/min and D = 3 mm. In the images, the dark part corresponds to the sample flowing downward through the packed bed. From the figures, it is found that the temperature in the reactor, i.e., the viscosity of the sample, and the diameter of packed materials affect the flow behaviors. The sample spreads to the radial direction as the temperature decreases or the diameter of particles in the reactor decreases.

CONCLUSIONS: In this work, it was revealed that the visualization of flow behaviors of heavy oils in the packed-bed reactor at high temperature and high pressure is feasible by neutron radiography.

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Figure 1 Schematic diagram of the apparatus



Figure 2 Neutron radiographs of flow behaviors of bitumen through the packed-bed.

PR2-4 Estimation of the Frosting and Defrosting Phenomena by Using Netron Radiography

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INTRODUCTION:

Heat exchangers which are operated at below 0 °C are subjected to the frost deposition and its growth. Frost formation on heat exchangers has a serious impact on the heat transfer performance in two ways; one is a thermal resistance between the cold surface and the humid ambient air, the other is an air-side pressure drop by the blockage effect of the frost layer. In spite of the relatively large number of articles, there are still gaps between the analytical model and the experimental results. One of the reasons is the difficulty to evaluate the mass transfer coefficient for the frost deposition.

On the cooled surface of a heat exchanger, the frost grows with a spatial distribution that is determined by the temperature and humidity profiles of the air and the flow pattern in the heat exchanger. This means that the mass transfer coefficient for frost formation has both temporal and spatial distribution. However, in conventional methods of measuring the frost accumulation rate, the frost is scraped from the surface, or the frost accumulation is estimated from the difference between the absolute humidities before and after the heat exchanger. In the present study, the frost formation and defrosting process was estimated by the two ways by neutron and X-ray radiographies at KUR B-4 radiation port.

ESTIMATION DEFROSTING PROCESS BY NEUTRON RADIOGRAPHY:

The frost layer with defrosting process was measured by using a neutron radiography in the defrosting process on the aluminum finned-tube heat exchanger under the forced-convection. The heat exchanger is shown in Fig.1. Fins have dimensions of 60mm in height, 28mm in width and 0.12mm in thickness.

The water deposition distributions were estimated quantitatively in every 5 seconds for 10 minutes of the defrosting period. The melting water behavior was investigated by comparing the water deposition distribution before defrosting. The initial stage of the defrosting was referred to as the melting progressing period, in which the melting water penetrated into the water permeation layer in the frost thickness direction, as shown in t=140sec. of Fig.2. After 180sec from starting the defrosting process, the frost at the fin front edge and the surrounding the tubes were melted, then, the water permeation layer was saturated by the melting water. The neutron radiography images, as shown at t=380 sec. of Fig.2, indicated the melting water penetration into the spanwise direction of the fins. The total migration volume of the melting water into the spanwise direction was about 30% of the total frost deposition.

MEASUREMENT OF FROST DENSITY PRO-FILE BY X-RAY RADIOGRAPHY:

Frost is the porous material of the crystal ice and air. The low thermal conductivity of the frost causes the low heat transfer performance. The frost density profile of the frost layer is the important property for the frost formation estimation. The frost formation process was measured by the X-ray radiography (XRG) on the cylindrical cooling surface.

The frost density profiles is shown in Fig. 3 at the 22.5mm from the reading edge of the cylindrical cooling surface. The experimental results clearly shows the development of the frost layer density.

CONCLUSIONS:

The neutron and X-ray radiographies are effectiveness for the frost layer measurement.



Fig.1 Plate-fin tube heat exchanger.



t = 380 sec.

(a) Digital camera image (b) Water deposition m_wFig.2 Water deposition profiles on the fins.



Fig.3 Distribution of frost density with height.

PR4 Project Research on the Elucidation of Generating Mechanism of Damaged Protein Induced by Aging and Irradiation

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Objectives and Allotted Research Subjects:

The aim of this project research is to elucidate the correlation between the change of the protein structure induced by various post-translational modifications with UV irradiation, gamma-irradiation, aging and protein function. We also investigate the repair mechanism for the damaged protein by irradiation. This research program has started in 2013. In this year, the 6 research subjects were carried out. The allotted research subjects (ARS) are as follows;

ARS-1: Detection of D-aspartyl endopeptidases activity in the Ascidian (*Ciona intestinalis*). (T. Kinouchi and N. Fujii)

ARS-2: Damage to biological molecules induced by ionizing irradiation and biological defense mechanisms against ionizing radiation II (T. Saito, I. Kim, T. Kanamoto and N. Fujii)

ARS-3: Separation condition of the peptides containing isomerized Asp residues after treatment of protein L-isoaspartyl methyltransferase (PIMT) (Y. Sadakane and N. Fujii)

ARS-4: Analysis of environmental stress-related hearing loss in mice and humans (N. Ohgami and N. Fujii)

ARS-5: Identification of biologically uncommon β -aspratyl residues in proteins using LC-MS/MS (N. Fujii, S. Kishimoto and N. Fujii)

ARS-6: D- β -Asp containing peptides are resistant to peptide bond cleavage compared with L- α -Asp containing peptides (K. Aki, E. Okamura)

ARS-7: One-shot LC-MS/MS analysis of deamidation of rat lens α - and β -crystallins induced by γ -irradiation (N. Fujii, I. Kim, T. Saito and T. Kanamoto).

Main Results and Contents of This Project

<u>ARS-1:</u> Kinouchi *et al.* detected D-Aspartyl endopeptidase (DAEP) activity in an experimental aquatic animal, the ascidian (Ciona intestinalis). The high DAEP activity in the ascidian was distributed in its reproductive tissues, but was quite low or not detectable in other organs. Since this activity was strongly inhibited by a synthesized DAEP inhibitor, it was suggested that the ascidian DAEP belongs to the same taxonomic family of the mammalian DAEP.

<u>ARS-2</u>: Saito *et al.* have irradiated rat lens tissue with 5 Gy γ -rays and determined the specific sites of the resulting oxidized amino acid residues (Cys 19, Cys 42, Trp 43, Met 44, Trp 69, Met 70, Met 102, Trp 157, Met 160, and Met 171) in γ E and/or γ F-crystallin.

<u>ARS-3</u>: Sadakane *et al.* prepared a repair enzyme for aged proteins, protein L-isoaspartyl methyltransferase (PIMT) by E. coli expression system, and determined the HPLC condition for separation of the peptides containing isomerized Asp residues generated after treatment of repairing enzyme, PIMT. We became able to determine easily the kinetics of cyclization and de-cyclization of succinimide intermediate.

<u>ARS-4</u>: Ohgami *et al.* suggested that barium levels in biological samples could be a new risk factor for hearing loss, especially at high frequencies of 8 and 12 kHz, in humans. We are investigating whether barium-mediated hearing loss involves aggregation of a specific protein in the auditory neurons in inner ears.

<u>ARS-5</u>: Fujii *et al.* established a new method to identify the β -Asp containing peptides by the analysis of the 2nd generation product ion spectrum of liquid chromatography - tandem mass spectrometry (LC-MS/MS).

<u>ARS-6</u>: Aki *et al.* demonstrated that D- β -Asp in α -crystallin mimic peptide was less active to peptide bond cleavage than L- α -Asp by using real-time NMR.

The trans conformer and low pKa of D- β -Asp side chain are thought to be the reason of less reactivity to peptide bond cleavage.

<u>ARS-7</u>: Fujii *et al.* determined the deamidation sites in crystallins after exposure to 5, 50, and 500 Gy of irradiated rat lenses by one-shot LC-MS/MS. The deamidation sites in rat irradiated crystallins resemble those reported in crystallins from human age-related cataracts. Thus, this study on deamidation of crystallins induced by ionizing irradiation may provide useful information relevant to the formation of human age-related cataracts.

Detection and Tissue Distribution of D-Aspartyl Endopeptidases Activity in the Ascidian (*Ciona intestinalis*)

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INTRODUCTION: We are researching a unique protease: D-Aspartyl Endopeptidase (DAEP), which we first discovered in the liver of mammals [1]. In these high body-temperature and long life span animals, DAEP seems to physiologically serve as a kind of quality-control system against the abnormal protein which contains D-isomer of Aspartate (D-Asp). It is strongly suggested that formation of D-Asp residue in proteins would eventually trigger the protein misfolding. Actually, D-Asp is detected in misfolded proteins of the age-related diseases (i.e., cataract, prion disease and Alzheimer's disease), and the formation of D-Asp in proteins is potentially noxious for the normal protein turnover in our cells and tissues. However, in searching a distribution of DAEP in various living things, except for mammals and birds, the activity could not be detected in livers, but rather in the reproductive tissue. For example, the tissue distribution of DAEP activity in the African clawed frog, Xenopus laevis, was remarkable in its testis and unfertilized eggs but was imperceptible in its liver. The trend mentioned above has been also observed in other non-vertebrates. As a result of searching for DAEP activity in Japanese green sea urchin, Hemicentrotus pulcherrimus, its high DAEP activity was also detectable in their testes, ovaries and unfertilized eggs. We therefore supposed that the original and primitive physiological functions of DAEP might be associated with fertilization and/or oocyte maturation and started examining the distribution of DAEP in an experimental aquatic animal, the ascidian, Ciona intestinalis.

EXPERIMENTS: Material> Wild and mature ascidians, *C. intestinalis* (were kindly gifted by Dr. Reiko Yoshida and Dr. Yutaka Satou, Kyoto University) were collected and then maintained in indoor tanks of artificial seawater at 18°C at the Maizuru Fisheries Research Station of Kyoto University.

Sample preparation> All subsequent operations were performed at 4°C. Surgically removed organs of the ascidian: heart, ovary, testis, stomach, intestine and genital duct were individually cut into fine pieces, homogenized by a small homogenizing tool, BioMasher[®] II, in 3 volumes of homogenization buffer (20 mM Tris-HCl (pH 8.5), 150 mM NaCl, 1 mM EDTA), and then centrifuged

at 100 × g for 5 min. The supernatant was centrifuged at 15,000 × g for 10min. The precipitate was suspended in 200 μ l of solubilization buffer (20 mM Tris-HCl (pH 8.5), 150 mM NaCl, 0.25% CHAPS, 1 mM EDTA) and was sonicated for 30 sec by TOMY Ultrasonic Disruptor UD-200 (at output level 4 and 50% duty cycle). Sonicated samples were centrifuged at 15,000 × g for 10 min, and the supernatants were applied to the DAEP activity assay

Measurement of DAEP activity> We developed an assay system for DAEP activity using the synthetic D-Asp containing substrate, Succinyl-D-Aspartic acid α -(4-methyl-coumaryl-7-amide) (Suc-D-Asp-MCA) [1]. Supernatant of the above samples was mixed and incubated with 0.1 mM Suc-D-Asp-MCA and the assay buffer (10 mM Tris-HCl (pH 8.5), 200 mM NaCl, 3 mM MgCl₂) at 25°C. The fluorescence of aminomethylcoumarin liberated from Suc-D-Asp-MCA by DAEP was measured at $\lambda ex = 380$ nm and $\lambda em = 460$ nm.

RESULTS & DISCUSSION: The high DAEP activity in the ascidian was distributed in its ovary and testis, but was quite low or not detectable in other organs (Table). The stomach also seemed to have low DAEP activity. Since contents in the stomach consisted of the fed and undigested diatoms (Chaetoceros gracilis) which includes a large quantity of reddish-brown pigment, it was assumed that the observed data was not a result of the DAEP activity in the ascidian stomach, but caused by auto-fluorescence of the diatomaceous pigment. The DAEP activities in the ascidian ovary and testis were strongly inhibited by the addition of Zn²⁺ or a synthesized DAEP inhibitor (Benzyl-L-Arg-L-His-D-Asp-CH₂Cl) [1]-[2]. Considering this result with previous studies regarding the sensitivity of the mammalian DAEP against the inhibitor, it was suggested that the ascidian DAEP belongs to the same taxonomic family of the mammalian DAEP.

Table. Relative activity of DAEP in organs of the ascidian. Meaning of symbols in the table, -: not detectable, +: very low, +++: high, ++++: very high

Organs	Heart	Ovary	Testis	Stomach	Intestine	Genital duct
DAEP activity	-	+++	++++	+	-	-

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PR4-2 Damage to Biological Molecules Induced by Ionizing Irradiation and Biological Defense Mechanisms against Ionizing Radiation II

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INTRODUCTION:

Some organisms have biological defense mechanisms against ionizing radiation [1]. Investigating the details of damage induced by ionizing radiation to biological molecules such as protein, DNA, and lipids is essential to elucidate the defense mechanisms of these organisms. However, there have been very few reports on protein damage induced by ionizing radiation in vivo. Ionizing radiation decomposes water and generates free radicals and reactive oxygen species such as OH, O_2 , and H_2O_2 , which can oxidize biological molecules [2]. Crystalline lens is excellent tissue for investigation of protein oxidation induced by external stresses, because its protein composition is simple. In this study, we irradiated rat lens tissue with y-rays and analyzed the resulting oxidation of amino acid residues of y-crystallin, which is considered to play an important role in maintaining the transparency and refractive index of the lens [3], by liquid chromatography-tandem mass spectrometry (LC-MS/MS).

EXPERIMENTS:

γ-Irradiation to rat lenses:

Lenses isolated from male Wistar rats, 4-weeks old, in 50 mM sodium phosphate buffer (pH 7.4) and 150 mM NaCl were irradiated with γ -rays at a dose of 5 Gy and a dose rate of 2.8 Gy/h. γ -Irradiation was carried out at the Co-60 Gamma-ray Irradiation Facility of the Kyoto University Research Reactor Institute.

Extraction and enzymatic digestion of proteins from rat lenses:

 γ -Irradiated rat lenses were homogenized by ultrasonication in 50 mM sodium phosphate buffer (pH 7.4), 150 mM NaCl, 1 mM phenylmethylsulfonyl fluoride (PMSF), and 1 mM EDTA. The samples were fractionated into water-soluble (WS) and water-insoluble (WI) fractions by centrifugation at 12,000 × g for 20 min at 4°C. The WI proteins were dissolved in 8 M urea, 50 mM sodium phosphate buffer (pH 7.4), and 150 mM NaCl; the urea concentration was then diluted to less than 1 M in the same buffer (pH 7.4) and 150 mM NaCl. The WS and WI proteins were digested with trypsin for 17 h at 37°C at an enzyme-to-substrate ratio of 1:50 (w/w).

Liquid chromatography-mass spectrometry analysis of tryptic peptides:

Peptides resulting from trypsin digestion were separated by nano-flow HPLC. MS analysis was carried out by alternating between full MS and MS/MS scans. MS/MS scans were performed using the collision-induced dissociation mode with dynamic exclusion function.

RESULTS:

In order to identify γ -irradiation-induced oxidation sites of amino acid residues in γ -crystallin, WS and WI proteins from γ -irradiated and non-irradiated rat lenses were digested with trypsin, and the resulting peptides were analyzed by LC-MS. The mass of the peptides were measured and analyzed by MS/MS, and then all peptides were identified using the Proteome Discover 1.0 software attached to the LC-MS system. In general, Met, Trp, and Cys are the amino acid residues susceptible to oxidation in proteins, and these residues can be converted into methionine sulfone (+16 mass as +10), methionine sulfoxide (+32 mass as +2O), hydroxytryptophan (+16 mass as +10), and cysteine sulfenic acid (+16 mass as +10) or sulfinic acid (+32 mass as +20). γ -Irradiation at 5 Gy resulted in oxidation of Trp 69, Met 70, and Met 102 in γE and/or γF -crystallin in the WS fraction, and oxidation of Cys 19, Cys 42, Trp 43, Met 44, Trp 69, Met 70, Met 102, Trp 157, Met 160, and Met 171 in yE and/or yF-crystallin in the WI fraction. The oxidation of amino acid residues can cause structural and functional changes of proteins. The study on details of ionizing radiation-induced damage of biological molecules will provide useful information for investigations of the biological effects of ionizing radiation and the corresponding biological defense mechanisms.

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PR4-3 Separation Condition of the Peptides Containing Isomerized Asp Residues after Treatment of Protein L-Isoaspartyl Methyltransferase (PIMT)

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INTRODUCTION: The stereoconversion of aspartyl (Asp) residue arise through intramolecular rearrangement, such as *via* a succinimide intermediate. The native L-Asp residue in the protein was converted to the L-succinimide intermediate, and it is quickly hydrolyzed and produced the mixture containing L-Asp and L-isoAsp residue in a ratio of approximately 1:3. L-isoAsp residue in a ratio of approximately 1:3. L-isoAsp peptide bonds in aged proteins by transferring a methyl group from S-adenosylmethionine to a α -carboxyl group of L-isoAsp residue (Fig.1). In this study, we prepared PIMT by *E. coli* expression system, and determined the HPLC condition for separation of the peptides containing isomerized Asp residues generated after treatment of repairing enzyme, PIMT.



Fig. 1 The β -linkage isomerization and stereoinversion to D-form of aspartyl residue and PIMT repair system.

EXPERIMENTS: The recombinant PIMT protein was prepared by His-tag conjugated *E. coli* expression system, and the four isomerized peptides (HFSPEDLTVK) of crystalline (named as T10) were synthesized by using Fmoc amino acids. To be the substrate for PIMT, L-isoAsp was substituted for the aspartate residues. The T10 peptide bearing L-isoAsp residue was treated by PIMT for 60 min at 37 °C, and the reaction was stopped by addition of HCl. The reacted sample was analyzed by reversed-phase HPLC with acetonitrile/H₂O containing 15 mM phosphate buffer, detected by absorption at 215

nm. The peptides bearing methyl ester and succinimide were isolated and determined their molecular mass to identify the retention time of HPLC.

RESULTS: PIMT transfers the methyl group of S-adenosylmethionine onto a α -carboxyl group of L-isoAsp residue of T10 peptide, then resulted in generating the peptide bearing methyl ester. The methyl ester undergoes spontaneously and rapidly demethylation (30-60 min, half-time) to generate succinimide intermediate. The intermediate was hydrolyzed (3-20 hr, half-time) to produce the T10 peptide beating L-Asp or L-isoAsp residue. To determine the amount of isomerized peptides above, we prepared the mixture of the T10 peptides bearing L-Asp, L-isoAsp, D-Asp and D-isoAsp, and PIMT-reacted solution, which included the peptide bearing methyl ester and succinimide. The mixture consisted of six peptides was separated by reversed-phased HPLC with 15 - 16 % acetonitrile with various pH of 15 mM phosphate buffer, and we found that the separation was accomplished in the condition using the eluent 15 % acetonitrile with pH 3.0 of 15 mM phosphate buffer (Fig. 2). We became able to determine easily the kinetics of cyclization and de-cyclization of succinimide intermediate.



Fig 2 Separation of T10 peptides containing various isomerized Asp residue by standard HPLC. The mixture of six peptides were separated by reversed-phase HPLC with ODS column using the eluent 15 % acetonitrile with pH 3.0 of 15 mM phosphate buffer. β L: the peptide containing L-isoAsp residue, β D: D-isoAsp residue, α D: D-Asp residue, methyl: methyl ester, α L: L-Asp residue, Suc: succinimide.

PR4-4 Analysis of Environmental Stress-related Hearing Loss in Mice and Humans

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INTRODUCTION: Barium can be detected in tube well drinking water and foods including seaweeds or nuts. Thus, we eat or drink barium contained in drinking water and foods in our daily life [1]. However, it has not been recognized that the ingestion of barium contained in food and water can be a potential risk to our health. In a previous study, exposure to barium has been shown to impaire physiological functions including blood pressure [2]. Our previous study further showed that exposure of mice to barium caused increased level of barium in inner ears, leading to hearing loss [3]. At present, however, it is not cleared about an association between barium and hearing loss in humans. Therefore, this study performed an epidemiological study in order to investigate an association of barium levels in biological samples and hearing levels in humans.

EXPERIMENTS: The study was performed for 145 subjects aged from 12 to 55 years who agreed in written form to participate in hearing examinations as previously described [4]. In short, this investigation was performed using a self-reporting questionnaire on smoking, age, clinical history, weight and height of subjects. The procedures were explained and informed consent was obtained from all of the subjects. This study was ethically approved by Nagoya University International Bioethics Committee following the regulations of the Japanese government (approval number 2013-0070). Auditory thresholds at frequencies of 1, 4, 8 and 12 kHz were measured by pure tone audiometry (PTA). We measured barium levels in biological samples by the method previously described [3]. In short, biological samples were put into a 15 ml polypropylene tube with 3 ml of HNO₃ (61%). The sample tubes were incubated at 80°C for 48 hours and then allowed

cool to room temperature for 1 hour. Then, 3 ml of H_2O_2 (30%) was added to each tube and the tubes were incubated at 80°C for 3 hours. After the samples had been diluted with ultrapure water, the Ba level in each sample was measured by using an inductively coupled plasma mass spectrometer (ICP-MS; 7500cx, Agilent Technologies, Inc.) with a reaction cell for absence of ArCl ion interference.

RESULTS: Our results showed hearing impairments in subjects with high barium levels in toenails. Our univariate analysis also suggested associations of barium levels in hair with hearing levels (1, 4, 8 and 12 kHz) in subjects. Multivariate analysis with adjustment for age, sex, BMI and smoking revealed that barium levels in hair had significant associations with hearing loss at 8 kHz (OR = 4.8; 95% CI: 1.4, 17.7) and 12 kHz (OR = 15.5; 95% CI: 4.0, 79.5).

Conclusions: Our study suggests that barium levels could be a risk factor for hearing loss in humans as well as in mice [4]. Results obtained in our previous study suggested that barium administered by drinking water specifically accumulates in inner ears resulting in severe impairments of hearing with degeneration of inner ears in mice [3]. A previous study has shown that exposure to a toxic element can cause aggregation of proteins resulting in neurodegeneration [5]. Therefore, we will further investigate whether exposure to barium causes aggregation of a specific protein in inner ears to clarify a mechanism of degeneration of auditory neu-rons.

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Identification of Biologically Uncommon β-aspratyl Residues in Proteins Using LC-MS/MS

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INTRODUCTION: We recently developed a method for four Asp isomers ($L\alpha$ -, $L\beta$ -, $D\alpha$ -, $D\beta$ -Asp) at individual sites in proteins by liquid chromatography - tandem mass spectrometry (LC-MS/MS) system because the four Asp isomers containing peptides were eluted at the different time on the chromatogram [1]. However, to identify the four Asp isomers on the chromatogram, it is necessary to synthesize reference peptides containing the four different Asp isomers as standards. This is time-consuming. Last year, we reported a method for identifying the β -Asp containing peptides by detecting the specific 2nd generation product ions using an ion trap mass spectrometer. In this study, we developed an advanced method for identifying the β -Asp containing peptide using a LC-MS/MS.

EXPERIMENTS: The peptides containing four different Asp isomers were made using a Fmoc solid-phase chemistry. The synthetic peptides were human αA -crystallin sequence from residues 55 to 65 peptides (aAT6 peptide) four Asp containing different isomers; TVL (La-D)SGISEVR $(\alpha AT6-L\alpha),$ TVL(Lβ-D)SGISEVR $(\alpha AT6-L\beta)$, TVL(D α -D)SGISEVR (α AT6-D α), TVL $(D\beta-D)SGISEVR$ ($\alpha AT6-D\beta$).

Human lens was homogenized in buffer by ultrasonication and fractionated into water insoluble (WI) and water soluble fractions by centrifugation at 16,000 g for 20 min at 4 °C. The WI proteins were dissolved in 8 M urea, 50 mM Tris/HCl, pH 7.8, 1 mM CaCl₂, and then the urea concentration was diluted to less than 1 M. The denatured WI proteins were digested with trypsin for 17 h at 37 °C.

LC used a nano-flow HPLC system (Paradigm MS4, Michrom Bioresources). MS was performed on an ion trap system (LCQ Fleet, Thermo). The 1st and 2nd generation precursor ion of α AT6 peptides were selected the [M+2H]²⁺ ion at 588.5 m/z and the y₈ (DSGISEVR) fragment ion at 862.4 m/z.

RESULTS: Fig. 1 shows the 2nd generation product ion spectrum of α AT6 peptide containing four different Asp isomers. The specific fragment ions of 789.6 m/z (y₈-73) and 771.6 m/z (y₈-91) were observed in the spectrum of α AT6-D β and α AT6-L β . The y₈-91 and y₈-73 were observed in the 2nd generation product ion spectrum of β containing α AT6 peptide (Fig 1b, d), while these product ions were not observed for α -Asp containing α AT6 peptide (Fig 1a, c). These results indicated that the 2nd generation product ion spectrum of β -Asp containing peptide from the α -Asp containing peptide.



Fig. 1 Second generation product ion spectrums of four isomeric Asp residues in α AT6 synthetic peptides; a) α AT6-L α , b) α AT6-L β , c) α AT6-D α and d) α AT6-D β .

Fig. 2 shows the mass chromatogram using a LC-MS/MS. Fig. 2a shows the set of mass chromatogram (588.0-589.5 m/z) of four Asp isomers aAT6 synthetic peptides. Fig. 2b shows the set of mass chromatogram (789.0-790.0 m/z) of 2nd generation product ion spectrum of aAT6 synthetic peptide containing four Asp isomers. The peaks of α -Asp (L α , D α) containing α AT6 synthetic peptides were not observed by the mass chromatogram (Fig. 2b). Fig. 2c shows the set of mass chromatogram (588.0-589.5 m/z) of the aAT6 peptide obtained from trypsin digestion of human lens protein. Fig. 2d shows the set of mass chromatogram (789.0-790.0 m/z) of 2nd generation product ion spectrum of aAT6 peptide obtained from trypsin digestion of human lens protein. The peaks of a-Asp (La, Da) containing aAT6 peptide were not observed by the mass chromatogram (Fig. 2d).



Fig. 2 Mass chromatograms (588.0-589.5 m/z) of LC-MS (a) α AT6 synthetic peptides and (c) α AT6 tryptic peptide of lens protein. Mass chromatograms (y₈-73; 789.0-790.0 m/z) of second generation product ion of (b) α AT6 synthetic peptide and (d) α AT6 tryptic peptide of lens protein.

CONCLUSION: Biologically uncommon β -Asp containing peptides were identified by the analysis of the specific 2nd generation product ion of LC-MS/MS.

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PR4-6 D-β-Asp Containing Peptides are Resistant to Peptide Bond Cleavage Compared with L-α-Asp Containing Peptides

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INTRODUCTION: Natural L- α -Asp isomerize to the uncommon D-β-Asp form via a succinimide intermediate. D-β-Asp is responsible for the structural change of proteins or peptides, because D-isomers with different sidechain orientation and β-isomers which prolong main peptide bond can induce uncommon main chain struc-tures, to trigger the ab-normal unfolding or aggregation leading to a disease. D- β - Asp residues are accumulated in aged human-lens αA-crystallin [1] [2]. High re-sistance of D- β -Asp to biochemical reactions is predicted. However, no information has been pro-vided about reactions such as bond cleavage next to D-β-Asp. In this study, spontaneous peptide bond cleavage next to Asp was compared between Asp isomers, by applying real-time solution-state NMR to eye lens Aα-crystallin 51–60 fragment

(α A51-60), S⁵¹LFRTVLD⁵⁸SG⁶⁰ consisting of L- α and α B-crystallin 61-67 analog (α B61-67), F⁶¹D⁶²TGLSG⁶⁷ consisting of L- α - and D- β -Asp 58 and 62, respectively.

EXPERIMENTS: Peptide synthesis Isomers of $\alpha A51\text{--}60$ and $\alpha B61\text{--}67$ composed of L- $\alpha\text{-}$ and D-β-Asp residues were synthesized by Fmoc solid-phase chemistry using an automated solid-phase peptide synthesizer (Shimadzu PSSM-8). Real-time NMR measurement Real-time ¹H-NMR measurements were carried out on 400 MHz spectrometer (JEOL ECA400) equipped with a super-conducting magnet of 9.4 T. A high sensitivity probe (JEOL, NM40T10A/AT) for 10-mm diameter tube was used. About 3 mg of aA51-60 and aB61-67 consisting of L-aor D- β -Asp residue was dissolved in 4 ml of 50 mM acetate buffer/D2O (pD 4.0) and subject to NMR measurement at 70 °C. Free induction decays were accumulated at 512-1024 times, corresponding to 1-2 h intervals. Amounts of reactant and product peptides were quantified by using integral intensities of the respective NMR signals. For $\alpha A51-60$, the signals of Asp58 H_{α} and Ser59 H_{β} , for α B61–67 Asp62 H_{α} and Thr63 H_{γ} were used [3].

side-chain conformers The population of staggered side-chain conformers (trans , gauche+ and gauche-) of α A51–60 and α B61–67 composed of L- α - and D- β -Asp residues was evaluated from vicinal spin-spin coupling constants H_{α}-H_{β 1} ($J_{\alpha\beta1}$) and H_{α}-H_{$\beta2$} ($J_{\alpha\beta2}$) obtained by high-resolution ¹H NMR measurement at 10 °C [3] [4].

RESULTS: The reactivity of D-β-Asp residue to peptide bond cleavage was different from that of L- α -Asp in both peptides. When the cleavage reaction was treated as first-ordered. rate constants were estimated to be $(6.9\pm0.5)\times10^{-3}$ h⁻¹ for L- α -Asp58 and (2.2±0.4)×10⁻³ h⁻¹ for D- β -Asp58 in α A51–60. Similarly, the rate constants in α B61-67 were estimated (4.7±0.9)×10⁻³ h⁻¹ for L- α -Asp62 and (1.3±0.2)×10⁻³ h⁻¹ for D- β -Asp62 [3]. Uncommon D- β -Asp was less active to peptide bond cleavage than $L^{-\alpha}$ -Asp residue. We consider why such difference in reactivity was found in L- α - and D- β -Asp residues. One is the difference in side chain conformers between $L-\alpha$ - and $D-\beta$ -Asp isomers. The cleavage reaction proceeds as : (i) when the carbonyl carbon (C_{CO}) of Asp peptide bond is attacked by hydroxyl oxygen of carboxylic side chain (O_{COOH}), a cyclic anhydride intermediate is formed; (ii) another fragment is also cleaved as a result of intramolecular cyclization; and (iii) the fragment including C-terminal Asp is finally produced by hydrolyzing cyclic anhydride intermediate. D- β -Asp58 and D- β -Asp62 more than 50% of the Asp side-chain is trans. The trans conformer of D- β -Asp side chain is thought to be disadvantageous for such reaction pathway because the long distance between C_{CO} and O_{COOH} interferes with the attack of O_{COOH} on C_{CO} to initiate intramolecular cyclization. Second reason is that pK_a of α -Asp (pK_a 4.0) and β -Asp (pK_a 3.3) side chain carboxyl is different[5]. At pD 4 in this work, the carboxyl group of D- β -Asp side chain is more ionized than L- α -Asp. When the bond cleavage undergoes from unionized state of carboxyl, the ionized carboxylate in D- β -Asp side chain is disadvantageous for the cleavage reaction.

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PR4-7 One-shot LC-MS/MS Analysis of Deamidation of Rat Lens α- and β-crystallins Induced by γ-irradiation

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INTRODUCTION: The eye lens is a transparent organ that functions to focus light and images on the retina. The transparency and high refraction of the lens are maintained by the ordering of fiber cells and stablelong-lived proteins, comprising α -, β -, and γ -crystallins. Because the lens crystallins are long-lived proteins, they undergo various modifications including isomerization, inversion, deamidation, oxidation, glycation and truncation. These modifications are generated by ionizing irradiation, UV light and oxidative stress, and decrease lens transparency and ultimately lead to the development of age-related cataracts. Of the modifications, deamidation induces the negative charge into proteins and alter protein-protein interactions. There are many data on the deamidation sites of lens crystallins from age-related cataract, but there are few studies of the specific sites of deamidation in young rat lens crystallins that have been

subjected to γ -irradiation. Thus we demonstrate the deamidation sites in rat crystallins after exposure to 5, 50, and 500 Gy of irradiation by one-shot LC-MS/MS.

EXPERIMENTS:

Application of γ **-irradiation to rat lenses.** Rat (Wistar rats, 4-weeks old, male) lenses in 50 mM sodium phosphate buffer (pH 7.4), 150 mM NaCl were irradiated with γ -rays at a dose of 5, 50 Gy or 500 Gy and a dose rate of 33.8 Gy/h. γ -Irradiation was carried out at the Co-60 γ -ray Irradiation Facility of the Kyoto University Research Reactor Institute.

Preparation of lens proteins. Rat lenses were homogenized and fractionated into water soluble (WS) and water insoluble (WI) fractions by centrifugation. The WI proteins were dissolved in 8 M urea for 1 h and then the final concentration of urea was diluted to less than 1 M in a buffer pH 7.8. The WS and WI proteins were digested with trypsin for 17 h at 37 °C at an enzyme-to-substrate ratio of 1:50 (w/w). The resulting peptides are applied to LC-MS/MS according to the previous method and analyzed by Proteome Discoverer 1.0 software. [1].

RESULTS:

Since deamidation of Asn or Gln residues generates Asp or Glu residues with an increase of 1 Da in the molecular mass of the tryptic peptide, LC-MS/MS analysis is very useful for the analysis of the deamidated sites in proteins. Figure 1 shows a typical example of the deamidation of



the Asn103 in βA3-crystallin 96-109 peptide (WDAWSGS¹⁰³NAYHIER, $[M+2H]^{2+} = 1692$) after 5Gy irradiated samples. The mass of the b1-b7 ions of the Asn 103-containing peptide shows the theoretical mass, but the mass of the b8-b13 ions was 1 Da greater in the 5 Gy-irradiated samples. Similarly, the mass of the y1-y6 ions shows the theoretical mass, whereas that of all y ions after y7 (i.e., y7-y13) was 1 Da greater for the irradiated peptides. The inset of Fig. 1 shows the elution time of deamidated peptides on LC. The elution time of the deamidated peptide (42.71 min) is different from the non-deamidated peptide (41.03 min). These results clearly indicate that the Asn 103 residue of βA3-crystallin was deamidated to an Asp residue by exposure to more than 5 Gy of γ -irradiation. Similarly, we found many deamidated sites in α -, β -crystallins from WI fractions.

CONCLUSION: The deamidation sites in rat irradiated crystallins resemble those reported in crystallins from human age-related cataracts. Thus, this study on deamidation of crystallins induced by ionizing irradiation may provide useful information relevant to the formation of human age-related cataracts.

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PR5 Studies on Actinides and Fission Products Performed at the KURRI Hot Laboratory

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1. Objectives and Allotted Research Subjects

Studies on actinide and fission product nuclides with careful management are being more important for reprocessing, disposal, partitioning, and transmutation processes in the nuclear fuel cycle. Hot laboratory of KUR-RI is one of core facilities in Japan, in which various nuclides can be handled. This project enhances utilization of the KURRI hot laboratory by opening for fundamental and application studies related to radiochemistry, nuclear chemistry, environmental chemistry, geochemistry, and so on. Allotted research subjects are;

- ARS-1 Complexation of actinides with organic substances (T. Sasaki *et al.*).
- ARS-2 Solubility of actinide compounds in aqueous media (T. Kobayashi *et al.*).
- ARS-3 Leaching of actinides and FPs from fuel debris (N. Sato *et al.*).
- ARS-4 Neutron irradiation damage of vitrified waste matrices (T. Nagai *et al.*).
- ARS-5 Ligand exchange reaction of actinides in molten salts (A. Uehara *et al.*).
- ARS-6 Electrochemical behavior of uranium in pyroreprocessing system (Y. Sakamura *et al.*).
- ARS-7 Structural study of f-elements in molten halides (H. Matsuura *et al.*).
- ARS-8 Molecular dynamics simulation of uranyl ion in molten salts (N. Ohtori *et al.*).
- ARS-9 Fundamental study of fission products for trans-actinide chemistry (Y. Kasamatsu *et al.*).
- ARS-10 Isotope separation by using microreactor (R. Hazama *et al.*).
- ARS-11 Optical properties of molten aluminum halides (T. Goto *et al.*).
- ARS-12 Precipitation of f-element oxides in molten halides (H. Sekimoto *et al.*).
- ARS-13 Uptake of radiocesium and radiopotassium in plants (T. Ohta *et al.*).
- ARS-14 Quantitative analysis of radionuclides in seawater (T. Kubota *et al.*).
- ARS-15 Isotopic composition of radionuclides in environmental samples (Y. Shibahara *et al.*).
- ARS-16 Noble gas mass spectrometry of neutron irradiated geological samples (H. Sumino *et al.*).
- ARS-17 ⁴⁰Ar/³⁹Ar dating of neutron irradiated minerals and glasses (O. Ishizuka *et al*).
- ARS-18 Radiometric Ar-Ar dating of neutron irradiated

lavas (N. Hirano et al.).

ARS-19 Behavior of fission products in soil samples (S. Fukutani *et al.*).

2. Main Results and Contents

ARS-1, 2, 3, and 4 were performed in order to deepen the knowledge of nuclear waste management issues. In ARS-1, for understanding the randomly cross-linked heterogeneous and macromolecule properties of humic substances (HSs), the complexation of actinides with HS was studied. The HS samples irradiated by Co-60 gamma-ray source at KURRI were characterized by NMR measurement, and the dependence of dose rate on the chemical structure was clarified. ARS-2 studied on the solid phase transformation of Th(OH)₄(am: amorphous) at 363 K. The apparent solubility and particle size of the solid phase after aging were evaluated. ARS-5, 6, 7, 8, and 12 were performed with the viewpoint of pyrochemistry. In ARS-5, the solubility of uranium in various molten alkali chlorides was determined. It was found that the solubility of uranium does not depend on the melting point of molten salts but increases with ionic diameter of solvent alkali metal ion. ARS-6 studied on the behavior of Al³⁺ in LiCl-KCl eutectic melt in order to develop the separation technique of Al from Al-actinides intermetallic compounds. It was found that Al can be separated from U by using a Cu electrode in LiCl-KCl melts. In ARS-7, the electrochemical measurement of zircomium in molten LiCl-KCl has been carried out for the first step of the elucidation of electrochemical behavior containing zircomium. The results suggested that zirconium would be difficult to be selectively electoreduced in molten LiCl-KCl depending on the concentration of zircomium. In ARS-8, the Helmholtz free energy changes for the successive additional processes of chloride ion to uranyl ion in aqueous solutions of LiCl with various concentrations have been evaluated. The stability of uranyl ion in hydrate melts was then discussed. ARS-12 studied on the formation of Nd(II), Dy(II) and Pr(II) in CaCl₂-LiCl eutectic molten salt with the combination of the absorption spectroscopy and the galvanostatic electrolysis method. The results showed that Nd(II) was decomposed through the disproportionation reaction. ARS-10 studied isotope fractionation of Ca. A possible fractionation of ⁴⁸Ca/⁴⁰Ca was found. ARS-13, 14, 15, 16, 17, and 18 focus on environmental chemistry and geochemistry by using KUR.

3. Summaries of the achievements

New and characteristic chemical, kinetic, structural, and thermodynamic data for actinides and fission products were obtained. These new information encompass solid chemistry, molten salt and solution chemistry, as well as nuclear reactions of f-elements and FPs. Restart of the KUR operation is strongly awaited.

PR5-1 Study on Chemical Property of Gamma-irradiated Natural Organic Matters

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INTRODUCTION: Due to the randomly cross-linked heterogeneous and macromolecule properties of humic substances (HSs) as natural organic matter, in soils, sediments, and natural waters, the apparent formation constant of actinides with HS changes depending on the solution conditions such as pH, metal and HS concentrations, and ionic strength. When the HS in underground water contacts with a radioactive waste package at repository site, the chemical property of HS should be altered by gamma-irradiation and the ability of complex formation may be decreased. The HS samples irradiated by Co-60 gamma-ray source at KURRI were characterized by NMR measurement, and the dependence of dose rate on the chemical structure was discussed.

EXPERIMENTS: The Eliot soil humic acid (SHA, 1S102H) and Suwannee river fulvic acid (SFA, 1S101F) were purchased from International Humic Substance Society (IHSS). The proton dissociation constants have been reported. The brownish aqueous solution samples for Co-60 gamma-irradiation were prepared to be 10⁻³ eq/dm³ at a given alkaline pH condition. The irradiation dose were 10, 100 and 500 kGy by controlling the time of exposure. ¹H NMR spectrum was obtained at 600.17 MHz (JEOL JNM-ECA600 FT NMR spectrometer) with acquisition time of 1.45 sec, pulse delay time of 5 sec, and accumulation of 1000 scans, and ¹³C NMR spectrum was obtained at 600.17 MHz with acquisition time of 0.69 sec, pulse delay time of 10 sec, and accumulation of 24000 scans.

RESULTS: The blackish precipitation was observed in the SHA solution after 500 kGy irradiation, and the same irradiation achromatized the SFA solution, suggesting a different degradation process. As shown in Fig. 1, the proton NMR spectra of HS have broad peaks at the entire chemical shift range and are different from that of general and commercially available reagents such as acetic acid and EDTA. Based on the previous studies about natural hydrocarbon compounds [1-4], these peaks of ¹H were assigned to be some kinds of functional groups. The integrated intensity of peaks is summarized in Table 1. The fragment of aliphatic hydrocarbon Hy increased after 10 kGy irradiation, while that of the aromatic protons H_{Ar} decreased. Meanwhile, the S/N ratio of ¹³C NMR spectra was significant low even in longer acquisition time. For better understanding of chemical property, a higher performance of NMR would need for further study.



Chemical Shift (ppm)



Table 1. Distribution% of protons in SHA based on the ¹H peak area at each range of chemical shift.

Dose (kGy)	Proton assigned (δ, ppm)				
	$H\gamma$ (0-	Hβ (0.9-	Hα (1.6-	Ho-c (3.0-	HAr (6.0-
0	7.8	16.7	23.3	33.3	18.9
10	10.8	16.9	22.9	32.5	16.9

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Effect of Solid Phase Transformation on the Solubility Product of Thorium Hydrous Oxide at 363 K

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INTRODUCTION: In the repository systems for high-level wastes or spent nuclear fuels, decay heat emissions are transferred to the groundwater aquifers surrounding the waste packages. Although the actual operating temperature depends on the structure of the repository system, temperature conditions greater than 333 K are thought to simulate results of temperature evolution up to a thousand years [1]. For evaluating the effects of the possible temperature on tetravalent actinide (An(IV)) migration behavior under certain scenarios, such as early failure of the waste package, it is necessary to determine the temperature effect on An(IV) solubility.

Under geological disposal conditions, An(IV) solubility is primarily controlled by amorphous hydroxide solid phase (An(OH)₄(am)). However, An(OH)₄(am) is thermodynamically meta-stable, and it is converted to stable crystalline oxides (AnO₂(cr)) by heating and calcination. A transformation of An(OH)₄(am) into a solid phase with higher crystallinity has also been shown to occur in aqueous solutions at high temperatures [2]. These observations indicate that the thermodynamic description of An(IV) solubility under elevated temperatures requires clarification of a possible transformation of An(OH)₄(am) as a solubility-limiting solid phase.

In the present study, we focused on the solid phase transformation of $Th(OH)_4(am)$ at 363 K. Sample solutions in contact with $Th(OH)_4(am)$ were stored in a chamber controlled at the aging temperature of 363 K. The apparent solubility and particle size of the solid phase after aging at 363 K were then investigated.

EXPERIMENTS: A 0.3 M 232 Th stock solution diluted to obtain sample solutions, and pH value was adjusted by HClO₄/NaOH. The sample solutions were kept in an oven controlled at 363 K for given periods. After aging, the sample solutions cooled down to 298 K and the pH values measured. The apparent solubility was measured by inductively coupled plasma mass spectroscopy (ICP-MS; HP4500, Hewlett Packard) after ultrafiltration (3k - 100k Da filter, Millipore), and the solubility product was determined. The detection limit for ICP-MS was about 10^{-8} M. The solid phases after aging at 363 K were separated, and then investigated using XRD (RINT 2000, RIGAKU). The Scherrer equation was applied to the XRD patterns to determine the particle size of the solid phases.

RESULTS: Th(IV) solubility after heating at 363 K for 7 days started to decrease and the values after 41 days were

approximately 3 orders of magnitude lower than those of $Th(OH)_4(am)$. In the XRD spectra of the solid phases kept at 363 K, broad peaks corresponding to $ThO_2(cr)$ were observed, indicating a progress of the crystallization of initial $Th(OH)_4(am)$.

In Figure 1, trends of the solubility product for different Th solid phases are shown as a function of particle size [3]. A dotted line represents an estimated solubility product according to the Schindler equation [4]. The solubility product value obtained in this study seems to be consistent with general trends of the data, but is much lower than the values for Th(OH)₄(am, fresh) and Th(OH)₄(am,dried), in spite of similar particle sizes. The observed difference might be explained by considering the higher sensitivity of the solubility value on the size of the smaller solid particles, and by considering possible differences in the size distributions of solid particles prepared by different experimental methods



Fig. 1. The effect of particle size on the solubility product of $Th(OH)_4(s,363)$ compared with the different solid phases and predicted curves from the Schindler equation [3].

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Solubility of Uranium (VI) Oxides in High Temperature Molten Alikali Chlorides

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INTRODUCTION:

In the non-aqueous reprocessing process of spent nuclear fuels by the pyrometallurgical and the electrowinning methods, a spent fuel is dissolved into molten alkali metal chloride eutectic and uranium and plutonium ions are recovered as metal or oxide. In the present study, the solubility of uranium ions in various kinds of molten alkali chlorides was measured.

EXPERIMENTAL:

Uranium oxides were converted to U_3O_8 by heating at 1023 K for 4 hours before use. Chlorination experiments were carried out at 973 K under an argon atmosphere, in which humidity and oxygen impurity were continuously kept less than 1 ppm. Chlorine gas was purged for half an hour after U_3O_8 addition to the chloride melts. The solution phase was separated from undissolving solid phase by using a glass filter which was attached at the end of the tube. Argon gas was purged to exchange chlorine gas dissolving in molten salt phase before the phase separation. Uranium concentration in molten salt phase was measured by hydrogen peroxide process. Molten salts used were LiCl, CsCl, RbCl, LiCl-KCl, LiCl-RbCl, NaCl-KCl, NaCl-KCl, NaCl-KCl, NaCl-KCl, and NaCl-CsCl eutectics.

RESULTS:

After 3.0 g U_3O_8 was putted into 3.1g LiCl-KCl eutectic melt at 973 K, chlorine gas was purged through the melts for over 1 hour at 20 ml/min.



Fig. 1. UV-Vis absorption spectra of uranium samples used in chlorination experimental (concentration depencence).

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 U_3O_8 was oxidized and dissolved into molten salt phase by chlorine gas as Eq. (1).

 $U_3O_8 + 3 Cl_2 \rightarrow 3 UO_2Cl_2 + O_2 \quad (1)$

In order to analyse the valence of the uranium ions in the salts, UV-Vis absorption spectrum [1] was measured after the dilution of the salt by LiCl-KCl eutectic (Fig. 1). Absorption peak at 370 nm corresponding to UO_2^+ as well as the peak at < 330 nm corresponding to UO_2^{2+} were observed. The concentration of UO_2^+ formed was calculated to be < 5% of total uranium concentration. It is suggested that UO_2^{2+} was partially reduced by Ar gas due to the shift of the equilibrium potential in the melt as Eq. (2).

$$UO_2Cl_2 \rightarrow UO_2Cl + 1/2 Cl_2$$
 (2)

Concentration of uranium ion in molten salt phase was measured by hydrogen peroxide method. The solubility of uranium in various molten salt was plotted as a function of melting point of the salts as shown in Fig. 2. The solubility of uranium in LiCl-KCl eutectic was 15.3 mol%, which was rather lower than that in NaCl-CsCl eutectic (37 mol%), RbCl melt (34 mol%). It was found that the solubility in CsCl was the highest in the molten salt tested. It is suggested that the solubility of uranium was not depended on the melting point of the molten salts but increased with increase of ionic diameter of alkali metal ion.



Fig. 2. Solubility of UO_2^{2+} depended on the melting point of the alkali chlorides.

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A Study on the Separation of Aluminum from Actinides in LiCl-KCl Eutectic Melt **PR5-4**

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INTRODUCTION: Electrorefining in molten LiCl-KCl eutectic-based salts is the key separation step in the pyrochemical treatment of spent nuclear fuels. It has been reported that the Al cathode is suitable for collecting actinides in the form of intermetallic compounds with a high separation factor against rare-earth fission products [1]. However, how to separate actinides from Al becomes problematic because U cannot be separated from Al by the anodic dissolution of U-Al alloy [2]. In this study, the behavior of Al³⁺ in LiCl-KCl eutectic melt was investigated to develop the separation technique of Al from Al-actinides intermetallic compounds.

EXPERIMENTS: LiCl-KCl eutectic melt was contained by a high-purity Al₂O₃ crucible in the temperature range 723-773 K, where a working electrode of W, Ta, Fe, Ni or Cu wire (1 mm diameter), a counter electrode of Al rod (3 mm diameter), a Ag/AgCl reference electrode and a type-K thermocouple were placed. The Ag/AgCl elec-trode consisted of a Ag wire immersed in a LiCl-KCl eutectic salt mixture with 1 wt% AgCl, which was con-tained in a closed-end Pyrex tube. The potential of the Ag/AgCl electrode was 2.410 V against a Li metal elec-trodeposited on a Ni wire. The concentration of Al³⁺ in the melt was changed by adding KAlCl₄ (APL Engi-neered Materials, 99.9% purity). The Al³⁺ concentration was determined by taking samples from the melt fol-lowed by ICP-AES analysis. All the measurements were conducted in a highpurity Ar atmosphere glove box (H_2O , $O_2 < 1$ ppm).

RESULTS: It is known that aluminum chloride has high vapor pressure. Figure 1 shows that the Al³⁺ concentration gradually decreased with time, which is probably due to the evaporation of AlCl₃ or Al₂Cl₆ from the melt. The evaporation rates of Al at 773 and 723 K were evaluated to be 5 x 10^{-5} and 1 x 10^{-5} g/cm²/h, respectively when the Al^{3+} concentration was 0.15 mol%. Hence, Al^{3+} can be spontaneously removed from LiCl-KCl melts containing actinide chlorides. It is necessary to examine how long it takes to reduce the Al^{3+} concentration to a sufficiently low level.

Cyclic voltammograms (CVs) of various metal wire electrodes (W, Ta, Fe, Cu and Ni) were measured in Fig. 2 CVs of W (\$\phi1x5 mm), Ni (\$\phi1x5 mm)\$ and Cu (\$\phi1x7 mm)\$ LiCl-KCl eutectic melts with dissolved KAlCl₄ to investigate deposition behaviors of Al at the cathode. On the W electrode, as shown in Fig. 2, a cathodic current rises at about -1.0 V, which corresponds to the deposition of pure Al metal. The sharp anodic peak at -0.95 V is due to the dissolution of the deposited Al metal. The redox po-

Y. Sakamura, M. Iizuka, K. Uozumi, A. Uehara¹ and T. Fujii¹ tential of Al/Al³⁺ on the W electrode is consistent with the CV of Al metal previously obtained in a LiCl-KCl-UCl₃ melt [2]. There is no other distinct peak, which indicates that W hardly forms alloys with Al under the condition of this measurement. The deposition behaviors of Al on Ta and Fe were similar to that of W.

> The CV of Cu has a cathodic peak at -0.90 V and an anodic peak at -0.75 V, which correspond to the formation and decomposition of an Al-Cu alloy, respectively. The anodic peak at -0.65 V might be due to a different Al-Cu alloy with less Al activity. The anodic current rising at -0.40 V is due to the dissolution of Cu into the melt in the form of Cu⁺ ion. It is suggested that Al can be separated from U by using a Cu electrode in LiCl-KCl melts containing Al^{3+} and U^{3+} because of the underpotential deposition of Al. The CV of Ni has an anodic peak at -0.45 V, which corresponds to the decomposition of an Al-Ni alloy. Since the cathodic current for the underpotential deposition of Al is small, the formation reaction of Al-Ni alloy might proceed slowly though the Al-Ni alloy is thermodynamically more stable than the Al-Cu alloy.





electrodes in LiCl-KCl-KAlCl₄ (0.099 mol%-Al) with scan rate of 0.1 V/s at 724 K.

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PR5-5 Electrochemical Behavior of Zirconium in Molten Alkali Chlorides

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INTRODUCTION: After the Fukushima Daiichi Nuclear Power Plant Accident, the reactor core has been experienced by high temperature from strong decay heat and leaded to fuel debris. The fuel debris may be expected to be the mixture of nuclear fuel, zircalloy theath, austenite stainless steel vessel, B₄C control blade and cement stuctural material. Especially, fuel material fisrt has contacted to zircalloy and resulted in uranium zirocnium oxide solid solution (corium). For the completion of decomissioning process, these heavily fuel contaminated materials should be properly treated and stablized in some ways. One of ideal options is pyrometallurgical process, which has simplicity and compactness feature. Our proposed process consists to fluorination and molten salt electrolysis. In this process, uranium is expected to be selectively fluorinated, dissolved into molten salts and electro-reduced into metallic form. However, the behavior of zirconium in solid solution is unpredictable in each process, thus fluorination behavior of uranium - zircomium oxide has been firstly investigated [1]. Here, the electrochemical measurement of zircomium in molten LiCl-KCl has been carried out for the first step of the elucidation of electrochemical behavior containing zircomium.

EXPERIMENTAL: The molten salt was prepared by the mixing LiCl – KCl eutectic and $ZrCl_4$ powder (0.5w/o) (AAPL) in a quartz cell. Three electrodes were used for electrochemical measurement; working electrode: a tungsten wire in 1mm ϕ , counter electrode: a pyrographite rod in 3mm ϕ and reference electrode: a silver wire dipped to LiCl – KCl - AgCl molten salt in borosilicate glass. To avoid from the contamination of oxygen and moisture, all electrochemical measurements have been performed in an argon circulated glove box (Mbraun) and electroanalyser (Hokuto Denko). In this year, cyclic voltammetry has been carried out at 550 °C.

RESULTS and DISCUSSION: Figure 1 shows variated cyclic voltammograms depending on voltage sweep region. As shown in Fig. 1, large reduction current at $-1.13 \text{ V} vs. \text{ Ag}^+/\text{Ag}$ is attributed to reduction to zirconium metal, which is relatively close to the decomposition of molten LiCl-KCl bath (it starts at ca. -1.25 V). This fact implies that zirconium would be difficult to selectively electoreduced in molten LiCl-KCl depending on the concentration of zircomium. Figure 2 shows sweep rate dependence of cyclic voltammograms. With increasing sweep rate, electoreduction potential of zirconium is

negatively shifted but electrooxidation potential of zirconium is almost independent to the sweep rate. This trend is typical semi-reversible reaction, which also implies difficulty of selective reduction of zircomium in LiCl-KCl.

In the next step, to be widen the reduction potential difference between zirconium and one of the elements of bath, two direction have been proposed, 1) to change the alternative molten chloride bath (NaCl-KCl and NaCl-2CsCl) or 2) to add small amount of fluoride in LiCl-KCl bath.



Fig. 1. Cyclic voltammograms of LiCl-KCl-ZrF₄(0.5w/o). Sweep rate: 100 mV/s



Fig. 2. Sweep rate dependence of cyclic voltammograms of LiCl-KCl-ZrF₄(0.5w/o).

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INTRODUCTION: Recently, hydrate melts such as concentrated alkali halide aqueous solutions attract some attention as one of the novel candidates for solvents useful for reprocessing of spent nuclear fuels[1]. We performed molecular dynamics (MD) simulation of LiCl aqueous solutions containing uranyl ion and reported some results such as diffusion coefficients and average local structure around uranyl ion in the previous report[2]. Furthermore, we have analyzed coordination structure around uranyl ions in 14 M LiCl aqueous solution in more detail and compared the results with experimental results[3]. In this work, in order to discuss the stability of uranyl ion in hydrate melts, we have evaluated the Helmholtz free energy changes for the successive additional processes of chloride ion to uranyl ion in aqueous solutions of LiCl with various concentrations.

CALCULATION: MD calculation has been performed under *NVE* ensemble. The SPC/E model was used for water molecule. The other parameters were taken from the references cited in [3]. The water and uranyl molecules were constrained by SHAKE algorithm. The number of particles included in the simulation cell is shown in Table 1. Time step was 1.0 fs. The Helmholtz free energy changes have been evaluated by both free energy perturbation (FEP) method and thermodynamic integration (TI) method. The successive additional processes denoted by R1, R2, R3, and R4 are as follows:

UO_{2}^{2+}	$+ Cl^{-} \rightarrow UO_2Cl^{+}$	•	•	• R1
$\rm UO_2 Cl^+$	$+ Cl^{-} \rightarrow UO_2Cl_2$	•	•	• R2
UO_2Cl_2	$+ Cl^- \rightarrow UO_2Cl_3^-$	•	•	• R3
$UO_2Cl_3^-$	$^{+} \text{Cl}^{-} \rightarrow \text{UO}_2 \text{Cl}_4^{2-}$	•	•	• R4

RESULTS: Figure 1 shows the free energy change as a function of distance between uranyl ion and chloride ion, which compares the results from both the results. First of all, we can see both the methods give the same results within the statistical errors. The minimum around 3-3.5Å means the most stable position within the first coordination shell. The maximum around 4-4.5Å means the energetic barrier caused by the first coordination shell. Figure 2 shows free energy changes for the successive additional processes from R1 to R4, which suggests that the most stable ionic species including uranyl ion is $UO_2Cl_4^{2-}$, regardless of the concentrations.

Table 1 Number of particles included the simulation cell for aqueous solutions of LiCl containing uranyl ion and the density of the simulated systems.

5				
[Cl ⁻] / mol dm ⁻³	$d / \text{kg cm}^{-3}$	H ₂ O	Number of un $UO_2^{2+}CI_2^{-}$	nit Li ⁺ Cl [−]
14	1.301	782	1	280
8	1.173	926	1	158
3	1.067	1044	1	58
0	1.001	1000	1	2



Fig.1 Free energy change for the additional process of Cl^{-} to UO_2^{2+} (R1) as a function of the coordination distance.



Fig.2 Free energy change for the successive additional processes of CI^- to uranyl ion as a function of the coordination number of CI^- by TI method.

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PR5-7 Study of Isotope Separation of Strontium and Calcium via Chemical Exchange Reaction

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INTRODUCTION: Calcium is congener of strontium and easy to handle to check the isotope effects. By utilizing chemical separation method for calcium isotope effect in liquid-liquid extraction, we can evaluate the ratios between the field shift effect to the mass effect.

EXPERIMENTS: Isotopic enrichment occurs according to the following chemical exchange reaction: $40 = 2^{+}$ $48 = 2^{+}$ $40 = 2^{+}$ $40 = 2^{+}$ where

$\epsilon / \Delta M$	Method and system	Temp.	Ref.
$[\times 10^{-5}]$		(°C)	
51~17	Liquid-liquid extraction	20±0.5	[1]
(Sr)	(LLE) with dicyclohex-		
	ano-18-crown-6		
	(DC18C6).		
	2.3M-0.5M Sr in aque-		
	ous phase.		
0.31	Cation-exchange chro-	25±0.2	[2]
(Sr)	matography with Sr lac-		
	tate.		
0.29	Cation-exchange chro-	25±0.2	
(Sr)	matography with Sr ace-		
	tate.		
0.10	Cation-exchange chro-	25±0.2	
(Sr)	matography with Sr		
	chloride.		
100	LLE with DC18C6.	25	[3]
(Ca)	0.07M CHCl ₃		
0.5~0.4	Solid-liquid chromatog-	—(Not	[4]
(Ca)	raphy(SLC)with ion ex-	speci-	
	change resin	fied)	
	(Dowex50B)		
3.7	SLC with ion exchange		[5]
(Ca)	resin (Dowex1,50).		
5.3	SLC with ion exchange	—	[6]
(Ca)	resin (Dowex50W).		
11	SLC with ion exchange	—	[7]
(Ca)	resin (Dowex) with NH_4		
	α -hydroxyisobutyrate		
51	SLC with ion exchange	—	[8]
(Ca)	resin (ANKB-50) with		

	iminodiacetate		
13~87	SLC with ion exchange	24-82	[9]
(Ca)	resin (PK-1) with coun-		
	ter-current		
2~5.2	SLC with ion exchange	25	[10]
(Ca)	resin (Asahi LS-6)		
12	SLC with ion exchange	25	[11]
(Ca)	resin (AG50WX4)		
88	SLC with crown-ether	88	[12]
(Ca)	resin (18C6, dimethyl-		
	sulfoxide)		
45	SLC with crown-ether	35	
(Ca)	resin (benzo-18C6).		[13]
	9M HCl (0.8mmφ,1m)		
38	SLC with crown-ether	35	
(Ca)	resin (benzo-18C6).		
	9M HCl (0.8mmφ,200m)		
75	SLC with crown-ether	25	[14]
(Ca)	resin (benzo-18C6).		
	Methanol		
48	SLC with crown-ether	40	[15]
(Ca)	resin (benzo-18C6).		
	Ethanol/HCl mixed		
13~24	Ion Exchange Elec-	20,25,	[16]
(Ca)	tromigration	40	
150 *	LLE with DC18C6	20	This
(Ca)	0.07M CHCl ₃		work
175 *	LLE with DC18C6	2	This
(Ca)	0.07M CHCl ₃		work

Table 1.Unit mass enrichment factors of Sr and Caisotope separation.*: Preliminary

RESULTS: Unit mass enrichment factors of Sr and Ca isotope separation are summarized in Table 1. In general, a crown ether gives a large separation factor with more than ten times, compared with an ion exchange method and has an applicable ability of separation in reality. **REFERENCES:**

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Disproportionation Reaction of RE(II) (RE = Nd, Dy, Pr) in CaCl₂-LiCl Eutectic Molten Salt

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INTRODUCTION: Mutual separation of Nd, Dy and Pr is essentially important for the recycling of the neodymium magnet. Uehara et al. reported on the mutual separation of La and Nd utilizing the disproportionation reaction of Nd(II) to metallic Nd and Nd(III) in LiCl-KCl molten salt [1]. This technique seems quite attractive comparing with the conventional processes such as the ion exchange process and the solvent extraction process in terms of the environmental load. In this study, the formation of Nd(II), Dy(II) and Pr(II) in CaCl₂-LiCl eutectic molten salt was investigated with the combination of the absorption spectroscopy and the gal-vanostatic electrolysis method.

EXPERIMENTS: Regents of CaCl₂, LiCl, NdCl₃, DyCl₃, and PrCl₃ was weighed and inserted into a optical cell attached with a cylindrical quartz tube and melted at 700 °C to form RECl₃(RE = Nd, Dy, Pr)-CaCl₂-LiCl molten salt. The galvanostatic electrolysis at -10 mA was conducted with 3 electrodes; the working electrode was a tungsten wire of 1 mm diameter, the counter electrode and the reference electrode was a silver wire of 0.5 mm diameter which immersed was in 1 mol%AgCl-CaCl₂-LiCl molten salt held in a cylindrical pyrex glass tube. The counter electrode and the reference electrode was electrically contacted with the RECl₃-CaCl₂-LiCl molten salt through the thin pyrex glass film. All potentials in this paper were measured against the Ag/ mol%) AgCl(1 electrode. The absorption spectra of the RECl₃-CaCl₂-LiCl molten salt was measured by UV-Vis absorption spectrometer with the reported procedure [2].

RESULTS: The cathodic potential during the galvanostatic electrolysis was as follows; -1.67 V for the NdCl₃-CaCl₂-LiCl molten salt, -1.50 V for the DyCl₃-CaCl₂-LiCl molten salt and -1.80 V for the PrCl₃-CaCl₂-LiCl system. The absorption spectra shown in Fig. 1 (a) and (b) indicates that Nd(II) and Dy(II) formed during the galvanostatic electrolysis in the NdCl₃-CaCl₂-LiCl molten salt and DyCl₃-CaCl₂-LiCl molten salt, respectively. On the other hand, the existence of Pr(II) was not confirmed in the absorption spectra shown in Fig. 1 (c). Consequently, the following reactions proceeded during the galvanostatic electrolysis.

$Nd(III) + e \rightarrow Nd(II)$	(1)
$Dy(III) + e \rightarrow Dy(II)$	(2)
$Pr(III) + 3e \rightarrow Pr$	(3)

As shown in Fig. 1 (a), the absorption peak attributing Nd(II) diminished in 10 min. after the electrolysis, which

indicates that Nd(II) decomposed through the disproportionation reaction.



Fig. 1. Absorption spectra before, during and after the galvanostatic electrolysis at -10 mA for (a) NdCl₃-CaCl₂-LiCl molten salt, (b) DyCl₃-CaCl₂-LiCl molten salt and (c) PrCl₃-CaCl₂-LiCl molten salt.

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Project Research on Nuclear Spectroscopy and Condensed Matter Physics Using Short-Lived Nuclei

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Objective and Participating Research Subjects

The main objectives of this project research are the investigation of the nuclear structure of unstable neutronrich nuclei and also the local properties of matters using short-lived nuclei.

This period is the second year of the project. Unfortunately, as in the last period, no experiments in all five research subjects of the project (27P6) were executed owing to the suspension of the reactor operation. Here, we report some results which were obtained in previous periods and have already been published in journals or submitted for publication.

The reports presented here are as follows:

Report 1. Thermal Stability of Impurities Ba and La at the Substitutional Sites in Fe

Report 2. Thermal Stability of Cd and In Impurities in ${\rm Fe}_3{\rm O}_4$

Report 3. Formation Process of Oxygen Vacancy in Zinc Oxide

Main Points Described in the Reports in the Following Three Pages

The experimental techniques used to measure hyperfine fields are in common: γ - γ -rays time-differential perturbedangular-correlation (TDPAC) technique. The nuclear probes and matters in which the probes are incorporated are (Report 1) ¹⁴⁰Ce arising from ¹⁴⁰Cs through ¹⁴⁰Ba and then ¹⁴⁰La (for short, ¹⁴⁰Ce($\leftarrow^{140}La\leftarrow^{140}Ba\leftarrow^{140}Cs$)) and Fe, (Report 2) ¹¹¹Cd($\leftarrow^{111}In$) and ¹¹¹Cd($\leftarrow^{111}In$) and ¹⁰⁰ ppm Al-doped ZnO.

Report 1 describes thermal stability of Ba and La at the substitutional sites in Fe. Although neither Ba nor La atoms (also Ce atoms do not) form a solid solution with Fe (therefore, the only possible method for the incorporation of these atoms in Fe is ion implantation), which implies that these atoms bond with Fe atoms very weakly, these atoms at the substitutional sites do not move at temperatures at least as high as 673 K. This result was obtained in the condition that atomic vacancies produced in the ion implantation are hindered to move and bond with Ba or La. The pre-implanted He atoms play a role in keeping the atomic vacancies to stay.

The aims of implanting ¹⁴⁰Cs in Fe using the KUR-ISOL (Isotope Separator On-Line) were the following two things: the first thing is to measure the nuclear magnetic moment of the first 4⁺ excited state of ¹⁴⁰Ce having a neutron magic number and to compare it with the nuclear theory. The second thing is to study the state of impurities He and Ce in Fe experimentally. This knowledge is

important in materials science related to nuclear fusion reactor materials. The results are already reported in journals.

Report 2 describes the locations of Cd and In impurities in magnetite, Fe_3O_4 , and their thermal stability, the details of which will be published in a journal.

About three decades ago, magnetic hyperfine field at ¹¹¹Cd(\leftarrow^{111} In) chemically doped in Fe₃O₄ was measured to study the supertransferred magnetic field from the magnetic (Fe) ions via the oxygen ions to the nonmagnetic (Cd) ions. The location of In was assigned to the tetrahedral (*A*) site. However, quite recently, a paper was published stating that the location of In was the octahedral (*B*) site. W. Sato *et al.* succeeded in obtaining TDPAC spectra with damping free for the first time, concluded that the location of In is the *A* site, and moreover obtained a signal indicating that at high temperatures some In ions in the A sites move to the B sites, which is consistent with a density functional theory calculation.

Report 3 describes an experimental method to obtain the formation energy of oxygen vacancy in ZnO, which is an important physical quantity because oxygen vacancies can bring about, for example, ferromagnetism in ZnO. S. Komatsuda *et al.* obtained the temperature dependence of the rate constant for ¹¹¹In migration in high-temperature vacuum from Al aggregations in 100 ppm Al-doped ZnO to the substitutional defect-free Zn sites. Considering the migration of ¹¹¹In as being induced by the formation of oxygen vacancies, they obtained 0.72(6) eV as the formation energy of oxygen vacancy, which is consistent with theoretical calculations.

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INTRODUCTION: Previously, we projected 100-keV ¹⁴⁰Cs ions into Fe foils at KUR-ISOL and observed, using the time-differential perturbed angular correlation (TDPAC) technique on the 2083-keV state of ¹⁴⁰Ce, that about 35% of ¹⁴⁰Ce arising from the β^- decay of ¹⁴⁰Cs through ¹⁴⁰Ba and then ¹⁴⁰La occupy the substitutional sites with no lattice defects nearby [1, 2]. The half-lives of ¹⁴⁰Cs, ¹⁴⁰Ba and ¹⁴⁰La are 63.7 s, 12.8 d and 40.3 h, respectively. About a week after the end of the ¹⁴⁰Cs bombardments, when the TDPAC measurements were started, all ¹⁴⁰Cs had decayed out and the number ratio of ¹⁴⁰Ba to ¹⁴⁰La was constant, 0.85, calculated from the values of their half-lives. We think that those ¹⁴⁰Ba and ¹⁴⁰La leading to ¹⁴⁰Ce located at the substitutional sites also occupy the same substitutional sites. Since neither Ba nor La atoms (also Ce atoms do not) form a solid solution with Fe, it might be expected that they easily move into different places with increasing temperature and are finally segregated. This time, we report on the thermal stability of impurities Ba and La at the substitutional sites.

EXPERIMENTS: Two Fe samples were prepared: one is Fe irradiated with 100-keV ¹⁴⁰Cs⁺ at KUR-ISOL and the other is Fe irradiated with 4-keV He⁺ using an ion beam gun and subsequently with 100-keV ¹⁴⁰Cs⁺. We name the first sample ¹⁴⁰Ce<u>Fe</u> and the second ¹⁴⁰Ce<u>He-doped Fe</u>. For these two samples, 0.5-h annealing in vacuum was performed at a same time in a same furnace at 473, 673, 773, 873, 973 and 1073 K in this order. Before and after each annealing, room-temperature TDPAC spectra of ¹⁴⁰Ce were obtained with two measurement systems, each consisting of standard fast-slow electronic modules and four BaF₂ scintillation detectors. The relevant cascade γ rays are the 329–487-keV ones and the angles between the cascade γ rays are 90° and 180°.

RESULTS: Fig. 1(a) and 1(b) show the TDPAC spectra for ¹⁴⁰Ce<u>Fe</u> and ¹⁴⁰Ce<u>He-doped Fe</u>, respectively. The oscillation pattern seen in some of the spectra is due to a unique static magnetic hyperfine interaction at ¹⁴⁰Ce occupying the Fe substitutional site with no lattice defects nearby [1]. Hereafter, we call these ¹⁴⁰Ce "good" ¹⁴⁰Ce. From the oscillation amplitude, "good" ¹⁴⁰Ce are about 30% and 40% of the implanted ¹⁴⁰Ce, respectively, for unannealed ¹⁴⁰Ce<u>Fe</u> and ¹⁴⁰Ce<u>He-doped Fe</u> ("35%" in INTRODUCTION is the average of these two values). We now look at Fig. 1(a) downwards. The oscillation amplitude gets reduced by about 20% at the annealing temperature of 473 K and about 50% at 673 K. At the annealing temperature of 773 K, there is no noticeable oscillation component, i.e., there is no "good" ¹⁴⁰Ce in ¹⁴⁰Ce<u>Fe</u>. We may consider that with increasing annealing temperature, parent atoms (Ba and La) at the substitutional sites move into different places producing various hyperfine fields at the ¹⁴⁰Ce, and/or that with increasing temperature, atomic vacancies migrate and bond to still parent atoms of "good" ¹⁴⁰Ce, producing various hyperfine fields at the ¹⁴⁰Ce. As explained below, the latter explanation is adequate at least below a certain temperature (we call it T_b) between 673 and 773 K.

Fig. 1(b) shows that all "good" ¹⁴⁰Ce remain "good" in ¹⁴⁰Ce<u>He-doped Fe</u> until at 673 K and about 60% of them do not at 773 K. This observation means that parent atoms (Ba and La) at the substitutional sites do not move at least below T_b . We think that the pre-deposited He atoms form He–vacancy clusters with vacancies mainly produced in the He and ¹⁴⁰Cs bombardments and that at annealing temperatures higher than T_b , He and vacancies are released from those clusters, migrate and bond to parent atoms of "good" ¹⁴⁰Ce. This interpretation is consistent with relevant results of a thermal helium desorption spectroscopy experiment (ref. 6 in [2]).

A closer look at Fig. 1(b) shows that the oscillation amplitude increases with annealing temperature up to 673 K: at 673 K it is larger than by about 18% that before annealing. We need to examine this point by carrying out additional experiments with various doses of He, which may also elucidate the reason for the difference in the abundance of "good" ¹⁴⁰Ce between the two samples before annealing.



Fig. 1. Room-temperature TDPAC spectra for (a) $^{140}Ce\underline{Fe}$ and (b) $^{140}Ce\underline{Fe}$ and (c) $^{140}Ce\underline{Fe}$, subjected to no annealing, 0.5-h annealing in vacuum at 473, 673, 773 and 1073 K (after annealed at 873 and 973 K).

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INTRODUCTION: Magnetite (Fe_3O_4) is one of the most promising magnetic compounds applicable to spintronic devices due to its excellent electric conductivity and magnetic property. In addition to the intriguing nature of pure Fe_3O_4 , it has been demonstrated that the physical properties of the oxide vary with the introduction of impurity ions, which allows us to expect wider application of the doped oxide.

In this respect, our interest was directed to functions of nonmagnetic indium ions (In^{3+}) in Fe₃O₄. The hyperfine fields at the impurity site have been reported in many papers in the past three decades; however, there is still a controversy even over their residential sites [1-3]. It is thus of importance for a basic study of solid-state physics of the compound to clarify the site(s) occupied by the impurity In ions.

In the present work, we succeeded in the preparation of ¹¹¹In-doped Fe₃O₄ exhibiting perturbed angular correlation (PAC) spectra with distribution-free Larmor frequencies composed of a unique component, and the impurity In site was unequivocally determined by a comparative study with the assistance of a different PAC probe ¹¹¹Cd(\leftarrow ^{111m}Cd). In addition to the site information, we successfully detected thermal behavior of In ions at high temperature. The element-dependent thermal stabilities observed for Cd and In are discussed.

EXPERIMENTS: About 3 mg of cadmium oxide (CdO) enriched with ¹¹⁰Cd was irradiated with thermal neutrons at Kyoto University Reactor to produce ^{111m}Cd. The neutron-irradiated CdO powder was then added into Fe_3O_4 powder and the homogenized mixture in the form of a disk was sintered at 1173 K for 45 min.

Commercially available ¹¹¹In HCl solution was added in droplets onto a disk of Fe_3O_4 . It then underwent heat treatment in a vacuum quartz tube at 1373 K for 3 h for the diffusion of the radioisotope.

TDPAC measurements of the ¹¹¹Cd(\leftarrow ^{111m}Cd) and ¹¹¹Cd(\leftarrow ¹¹¹In) probes were performed for the synthesized samples at various temperatures.

RESULTS: The TDPAC spectra of ^{111m}Cd(\rightarrow ¹¹¹Cd) and ¹¹¹Cd(\leftarrow ¹¹¹In) embedded in Fe₃O₄ are shown in Fig. 1. The Larmor frequencies ω_L observed for both probes at room temperature show good agreement with each other,

signifying that both probes reside in the same site. At high temperature above T_C , however, contrastive patterns appear in their spectra. For the 111mCd probe, the directional anisotropy is unperturbed during the time of the present observation. This observation clearly shows that the probe nucleus is surrounded by a charge distribution of cubic symmetry; that is, ^{111m}Cd ions initially occupy the tetrahedral A site. This is also true of the ¹¹¹In ions because of the same ω_L values at R. T. However, the different observation for the different probes at high temperature implies that the ¹¹¹In ions migrate into another site. The spectrum in Fig. 1(c) was well fitted assuming two different components: the major component is unperturbed and the minority has an axially symmetric electric field gradient. Detailed investigation of the temperature dependence of PAC spectra revealed that the minor component corresponds to the probes at the substitutional B site [4]. In addition, DFT calculations suggest that the thermal stability of In ions are lower at the A site than that of Cd [4]. From these observations, we conclude that Cd ions are stabilized in the A site at least in the present temperature range, while In ions at the A site jump out of the potential at high temperature to move into the *B* site. We believe that the thermally activated displacement of In ions could be an important clue to materials design for impurity-doped magnetite.



Fig. 1. PAC spectra (a) of ${}^{111}Cd(\leftarrow {}^{111m}Cd)$ at 873 K, (b) of ${}^{111}Cd(\leftarrow {}^{111m}Cd)$ at R. T., (c) of ${}^{111}Cd(\leftarrow {}^{111}In)$ at 900 K, and (d) of ${}^{111}Cd(\leftarrow {}^{111}In)$ at R. T.

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INTRODUCTION: Defect-induced properties of zinc oxide (ZnO) have been attracting much attention toward their application to functional materials in a wide field of industry. Especially, physical properties brought about by Al ions and/or oxygen vacancies in ZnO are one of the most intriguing topics for the development of future electronic devices. Extrinsic-semiconductor devices such as of Al-doped ZnO are expected to be in use under various ambient conditions; the states of being of impurity ions in the matrix are susceptible to change depending on the condition. For a practical use of Al-doped ZnO device, therefore, we have investigated the local structures in Al-doped ZnO under various ambient conditions by means of the time-differential perturbed angular correlation (TDPAC) method. In one of our previous TDPAC studies, we observed contrasting atmosphere dependence of the stability of aggregations of ¹¹¹In and Al impurities doped in 100 ppm Al-doped ZnO: (i) Al and In impurities associate with each other by their thermal diffusion in air, and (ii) the ¹¹¹In probe is detrapped from the Al aggregations in high-temperature vacuum, resulting in substitution at defect-free Zn sites [1,2]. Detailed investigation of the thermal behavior of the impurities has revealed that the dissociation reaction is triggered by the formation of oxygen vacancies. In the present work, in order to extend quantitative discussion on the kinetics of the formation process of oxygen vacancy, we evaluated the formation energy of oxygen vacancy from the temperature variation of the detrapping process during heat treatment in vacuum [3].

EXPERIMENTS: For the synthesis of 100 ppm Al-doped ZnO, stoichiometric amounts of Al(NO₃)₃ · 9H₂O and ZnO powder were mixed in ethanol. The suspension was heated to evaporate the ethanol until dryness. The powders were pressed into disks and sintered in air at 1273 K for 3 h. Commercially available ¹¹¹In HCl solution was added in droplets onto the sintered disks. The initial concentration of ¹¹¹In ions doped in the sample was typically ~100 ppt. After the disks were dried up by heat, they again underwent heat treatment in air at 1373 K for 2 h. Following the doping of ¹¹¹In, each disk was separately sealed in different quartz tubes in vacuum, and 24-h isochronal annealing was performed one by one at different temperatures (873~1148 K). The TDPAC measurements were carried out for the 171-245 keV cascade γ rays of the ¹¹¹Cd(\leftarrow ¹¹¹In) probe with the intermediate state of I = 5/2 having a half-life of 85.0 ns.

RESULTS: It was found from the TDPAC spectra of the ¹¹¹In(\rightarrow ¹¹¹Cd) in 100 ppm Al-doped ZnO heat-treated at various temperatures that ¹¹¹In probes come to detrap from the Al aggregations in high-temperature vacuum by degrees. It was revealed that this detrapping process is controlled by the first-order rate law. For the first-order reaction of the dissociation process, we obtained the rate constant *k* at different temperatures. Their temperature variation is plotted in Fig. 1. A least-squares fit to the *k* values was then carried out with the following Arrhenius equation:

$$k = v_0 \exp\left(-E_a / k_{\rm B}T\right)$$

and the activation energy, E_a , was evaluated to be 0.72 (6) eV. Because the dissociation process of the ¹¹¹In probe and Al would be induced by the formation of oxygen vacancies as discussed above, we suggest that the observed E_a is closely related to the formation energy of oxygen vacancies in Al-doped ZnO sample. This interpretation is supported by the fact that the present E_a value shows good agreement with the theoretical ones calculated for the formation energy of oxygen vacancies in ZnO (0.8 and 1.0 eV)[4,5].



Fig. 1. Temperature dependence of the rate constant, k. An Arrhenius equation was used for the fit.

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OBJECTIVES AND PERFORMED RESEARCH SUBJECTS:

Mössbauer spectroscopy is a powerful and well established method for wide variety areas of researches, as physical-, chemical-, biological-, such and earth-sciences. As the Mössbauer resonance line is extremely narrow, hyperfine interactions are well resolved and give us the information on the surrounding electronic states and magnetism. The element specific information, which is one of the most superior features of the Mössbauer spectroscopy, is important and required for modern precise materials science and complex systems such as biological substances. Moreover, Mössbauer spectroscopy is useful and valuable because it usually does not demand the doping of radioactive sources for the measured samples; the decay of the unstable nucleus sometimes induces the change of local electronic states.

The main objectives of this project research are the investigation of the fundamental properties of new materials and the development of the advanced experimental methods by using multi-element Mössbauer spectroscopy under high-magnetic fields.

However, owing to the shutdown of the research reactor for the long term, some of the planned researches have not been performed.

The research subjects performed are as follows:

- P7-1 Mössbauer Spectroscopy in Applied Magnetic Field of Multiferroic Iron Oxides (S. Nakamura *et al.*).
- P7-3 Mössbauer γ-ray size focused using MCX measured by Si-PIN semi-conductor detector (K. Shinoda, *et al.*).
- P7-4 Mössbauer spectroscopy of Fe₆₅Ni₃₅ alloy irradiated by proton with 2 MeV (M. Matsushita, *et al.*).
- P7-6 Mössbauer Study of BaFe₂As₂ under High Magnetic Fields (S. Kitao, *et al.*).

MAIN RESULTS AND CONTENTS OF THIS REPORT:

The following reports were contributed by research groups in this project research.

In order to clarify the mechanism of the multiferroicity in iron oxides, S. Nakamura *et al.* (P7-1) studied h-ErFeO₃, which has ferroelectric ($T_c > 900$ K) and antiferromagnetic ($T_N = 120$ K) properties, by using ⁵⁷Fe Mössbauer spectroscopy. The Mössbauer spectra were measured at 4.2 K in an applied magnetic field of 5 T parallel to the c-axis. The ⁵⁷Fe Mössbauer measurements and the magnetization measurements suggest that the magnetic moment of Fe³⁺ has canted antiferromagnetic nature.

K. Shinoda *et al.* (P7-3) have measured the size of focused 14.4keV Mössbauer γ rays from a ⁵⁷Co source obtained by a multi-capillary x-ray lens (MCX) to evaluate the performance of Mössbauer micro-spectrometer which has been developed. They also measured the size of focused 6 keV Fe K- α x rays from the ⁵⁷Co source. To identify the energies, on the contrary to previous measurement, they used a Si-PIN semi-conducting detector to distinguish the each energy.

Irradiation of the energetic particles, such as ions, neutrons and electrons, enlarges the ferromagnetic property in Fe-Ni alloys of concentration ranges near 35 at% Ni with face centered cubic structure (FCC) to high temperatures although the detailed mechanism is not understood. M. Matsushita, *et al.* (P7-4) performed Mössbauer spectroscopic measurement of the proton irradiated Fe₆₅Ni₃₅ alloy in order to investigate the variation of the magnetic microstructure caused by the irradiated Fe₆₅Ni₃₅ alloys at 473 K show the difference, and this indicates the change of the local magnetic structures by the irradiation.

After the discovery of Fe-oxipnictide superconductors, several series of Fe-based superconductors have been successively discovered and extensively investigated to elucidate the mechanism of their superconductivity. Recently, in the isovalent doped $BaFe_2(As_{1-x}P_x)_2$, the electronic nematic phase was found near above the magnetic transition temperature. Since the electronic nematic phase is considered to have some relationship in mechanism of superconductivity, the investigation of this phase has a great importance. S. Kitao et al. (P7-6) detailed temperature dependences of measured Mössbauer spectra under high magnetic fields to reveal the magnetic properties of the parent compound, BaFe₂As₂. The measured temperature dependent spectra under magnetic fields imply that the nature of the nematic phase is not due to magnetic order but asymmetry in the structure, which is consistent with the initial finding.

Mössbauer Spectroscopy in Applied Magnetic Field of Multiferroic Iron Oxides

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INTRODUCTION: Recently multiferroic properties of iron oxides have been extensively investigated. In addition to the macroscopic investigation such as dielectric and magnetic measurements, the microscopic and dynamic investigations are required in order to clarify the mechanism of the multiferroicity. The 57Fe Mössbauer spectroscopy is one of the appropriate probes. The authors have already conducted Mössbauer spectroscopy on the multiferroics GaFeO₃ [1, 2] and CuFeO₂ [3, 4], and found the changes in the quadrupole splitting related to the multiferroicity. In the present investigation, we deal with other multiferroic iron oxides by Mössbauer spectroscopy in applied magnetic field. Here in this report, we describe the result of h-ErFeO₃. The material is ferroelectric ($T_c > 900$ K) and at the same time antiferromagnetic ($T_{\rm N}$ = 120 K). The magnetization measurements and the 57Fe Mössbauer measurements suggest that the magnetic moment of Fe³⁺ has canted antiferromagnetic nature [5, 6].

EXPERIMENTS: The single crystal of ⁵⁷Fe-enriched h-ErFeO₃ thin film deposited on Al₂O₃ (0001) substrate was used as an absorber. The ⁵⁷Fe Mössbauer spectroscopy was conducted in conventional transmission geometry by using ⁵⁷Co-in-Rh (50 mCi) as the γ ray source. The incident γ ray direction was parallel to the *c*-axis. The spectra were measured at 4.2 K in applied magnetic field of 5 T parallel to the *c*-axis. The Doppler velocity was calibrated by using Fe metal foil. Lorentzian line shapes were assumed for the analysis.

RESULTS: In Fig.1, Mössbauer spectra of h-ErFeO₃ at 4.2 K in applied magnetic field of 5 T is shown. Strong 2-5th line intensity indicates that the Fe³⁺ magnetic moments lie almost in the *c*-plane. Compared with the spectra without magnetic field, the spectral shape dose not change so much. This indicates that the canted antiferromagnetic structure is well retained even in applied magnetic field of 5 T. The outermost lines slightly split into two lines, and thus the spectra. The analyzed as superposition of two subspectra.

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observed field H_{obs} are 41.0 and 42.6 T, slightly smaller and larger than $H_{\rm hf}$ of 41.4 T obtained without magnetic field. Since H_{obs} is a vector sum of H_{hf} and the applied magnetic filed H_{ex} , the two subspectra correspond to the up- and down-domain magnetic moments, respectively. This feature seems to be consistent with the fact that the magnetization curve at 5 K dose not shows hysteresis up to 5 T. The Euler angle (θ , φ) of the hyperfine field ($H_{\rm hf}$) with respect to the EFG z-axis reveals that the Fe³⁺magnetic moments form a 120° structure in the c-plane but incline several degrees toward the c-axis. configuration well depicts This а canted antiferromagnetic structure. Noted that $e^2 q Q/2$ is -2.432 mm/s, which is larger than that without magnetic field, -2.156 mm/s. This suggests that the local structure around Fe^{3+} ion becomes further distorted by H_{ex} . Such distortion may be caused by a displacement of Fe^{3+} ion toward the c-axis, which can be an origin of the magnetoelectric effect of this material.



Fig. 1. Môssbauer spectra of h-ErFeO₃ at 4.1K in applied magnetic field.

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INTRODUCTION: In the 2014 KURRI Progress Report, we estimated focus size of 14.4keV γ -ray of Mössbauer microspectorometer using a multi-capillary X-ray lens (MCX). In that report, a proportional counter was used as γ -ray detector. In this report, a Si-PIN semi-conductor, of which energy resolution is better than a proportional counter, is used to fix the spot size of focused γ -ray by MCX. The spot size is estimated by scanning tungsten edges along horizontal and vertical directions using auto mapping stages (2014 KURRI Progress Report). 6keV FeK X-ray is also emitted from

 ^{57}Co $\gamma\text{-ray}$ source and focused as 14.4keV. FWHM of 6keV X-ray is also estimated.

EXPERIMENTS and RESULTS: 14.4keV γ -ray and 6keV X-ray intensity distributions are measured with a Si-PIN semi-conductor detector by scanning tungsten edges with 20µm step, 780s exposure per a step, over 2000µm ranges (101steps). Fig.1 and 2 show horizontal intensity distributions of 6keV X-ray and 14.4 keV γ -ray, respectively. Fig.3 and 4 show vertical intensity distributions of 6keV X-ray and 14.4 keV γ -ray, respectively. In KURRI Progress Report 2014, FWHM of 14.4keV was estimated to 400µm. FWHM of 6keV was 650µm. In this study, FWHM of 14.4 keV γ -ray is 515 µm along horizontal and 465 µm vertical orientations. FWHM of focused γ -ray measured with proportional counter is not different from that of a semi-conductor detector.



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INTRODUCTION: The ferromagnetic range in Fe-Ni alloys of concentration ranges near 35 at% Ni with face centered cubic structure (FCC) was expand to high temperature range due to the irradiation of energetic particles, such as, ions, neutron and electron [1-3]. However it have not been understood that the detail of irradiation-induced ferromagnetism. Therefore we have realized Mössbauer spectra of proton irradiated Fe₆₅Ni₃₅ alloy in order to investigate the variation of the magnetic microstructure caused irradiation.

EXPERIMENT: A $Fe_{65}Ni_{35}$ alloy ingot was made by Ar arc-melting. The ingot was homogenized at 1273 K

for one week in evacuated silica tube, and then quenched. The $Fe_{65}Ni_{35}$ foils with the thickness of 8 μm were made from the ingot by the mean of rolling. After rolling the alloy was annealed at 1273 K for 3 hours to remove the residual strain. The foils were irradiated at proton with 2 MeV. Mössbauer spectra of $Fe_{65}Ni_{35}$ alloy and that irradiated by proton were been measured at various temperatures.

RESULTS: The obtained Mössbauer spectra of $Fe_{65}Ni_{35}$ alloy and that irradiated by proton were shown in Figure 1. The shape of spectra confirmed above that obtained in 473 K is difference between non-irradiated and irradiated $Fe_{65}Ni_{35}$ alloy, which means that the local magnetic structure was changed by the irradiation.

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Figure 1. Mössbauer spectra of Fe₆₅Ni₃₅ alloy (a) and that irradiated by proton with 2 MeV (b).

PR7-4

Mössbauer Study of BaFe₂As₂ under High Magnetic Fields

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INTRODUCTION: After the discovery of the so-called "1111" series of Fe-oxipnictide superconductors, LaFeAsO_{1-x}F_x[1], several series of Fe-based superconductors have been successively discovered and extensively investigated to elucidate the mechanism of their superconductivity. Among the related Fe-based superconductors, the so-called "122" series has been found by hole doping (K-doping) of BaFe₂As₂[2]. The superconductivity emerges in "122" series by various doping not only with holes or electrons but also by isovalent doping (P-doping). Recently, in the isovalent doped $BaFe_2(As_{1-x}P_x)_2$, the electronic nematic phase was found near above the magnetic transition temperature[3]. Since the electronic nematic phase is considered to have some relationship in mechanism of superconductivity, the investigation of this phase has a great importance.

Since ⁵⁷Fe-Mössbauer spectroscopy is an essential method to extract the electronic states of Fe, a number of Mössbauer studies on these Fe-based superconductors have been carried out and revealed many important facts, for example, in LaFeAsO_{1-x} F_x [4]. Moreover, Mössbauer spectroscopy under high magnetic fields has advantageous to reveal the nature of magnetism in these compounds[5]. In this study, detailed temperature dependences of Mössbauer spectra under high magnetic fields were measured to investigate the magnetic properties of the parent compound, BaFe₂As₂.

EXPERIMENTS: BaFe₂As₂ was synthesized by heating of a stoichiometric mixture of ground Ba and FeAs in a crucible of aluminum oxide sealed in a quartz tube, as in the reported method[2]. The obtained polycrystalline powder was characterized by x-ray diffraction and magnetic susceptibility measurements. For BaFe₂As₂ the superconducting transition was not observed but the magnetic transition was observed at 140K. ⁵⁷Fe-Mössbauer spectra were measured using a pellet of powder sample using a ⁵⁷Co source in Rh matrix with a nominal activity of 1.85 GBq. The velocity scales are referenced to α -Fe. Magnetic fields were applied by a superconducting-magnet cryostat with a direction parallel to the γ -rays.

RESULTS AND DISCUSSION: Mössbauer spectra of $BaFe_2As_2$ under the magnetic transition temperature showed magnetically-split pattern as in Fig 1(a) at 2.5K. When high magnetic fields are applied, the direction of magnetic moments has changed partly towards the direction of applied magnetic fields as in Fig 1(b). The spectra were well understood by a model of antiferromagnetic powder under the magnetic fields. This fact is a good proof that the antiferromagnetic order has occurred at the

magnetic transition temperature.

On the other hand, Mössbauer spectra of BaFe₂As₂ above the magnetic transition temperature has not magnetically split pattern as shown in Fig. 1(a) above 140K. The spectra showed 4-line patterns under the magnetic fields as shown in Fig. 1(b). This pattern can be well understood as a paramagnetic feature, where the magnetic moments are entirely aligned to the direction of the applied magnetic fields. The spectrum at 160K, which is near above the magnetic transition temperature, showed the wider line width than spectra far above. This broadening of the line width is considered to be due to the electronic nematic phase. However, the line broadening has not appeared in the spectrum under the magnetic field. That is, the broadening may not be originated by internal magnetic fields but by quadrupole splittings. This fact implies the nature of the nematic phase is not due to magnetic order but asymmetry in the structure, which is consistent with the initial finding[3].

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Fig. 1. Typical Mössbauer spectra of $BaFe_2As_2$ (a) without magnetic field and (b) under magnetic fields of 10T.

Translational Research on BNCT for Clinical Application

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Background and Objective

Using Kyoto University Research Reactor (KUR), patients with malignant tumors greater than 500 have been treated with boron neutron capture therapy (BNCT). Malignant brain tumors and head and neck cancers have been main malignancies treated with BNCT. Our laboratory (Division of Particle Radiation Oncology) has investigated the possibilities for new applications for BNCT. According to promising results in pre-clinical study, we have already treated some patients with liver cancers with BNCT and carried out clinical study on phase I study on BNCT for malignant pleural mesothelioma (MPM).

Promising clinical results of BNCT using the research reactor encouraged us to go to further stage of BCNT using an accelerator-based (AB) BNCT system. Co-operation of Kyoto University Research Reactor Institute and Sumitomo Heavy Industry have developed AB BNCT system with compact cyclotron as an accelerator. In 2012 and 2014, clinical studies on BNCT for recurrent malignant brain tumors and head and neck tumors to get an approval as a medical device from the Pharmaceuticals and Medical Devices Agency (PMDA), a Japanese regulatory agency. In a transition period from reactor-based (RB) BNCT into AB-based BNCT, many research issues should be dissolved from impending and long-term viewpoints.

Main objectives of our project is to dissolve many impending clinical issues to perform BNCT safely in AB-BNCT system and to investigate many research projects for many patients with cancer to be treated with AB-BNCT system.

Research Subjects

To advance RB-BNCT into AB-BNCT, a lot of researchers in various research fields such as clinical radiation oncology, medical physics, pharmacology, boron chemistry, and accelerator engineering are needed to be involved in our research projects. In this viewpoint, this research project consists of three research subjects (RS) as follows,

RS1. Clinical studies on BNCT

RS2. Pre-clinical studies on physiological and pharmacological aspects of BNCT

RS3. Medical physics studies on BNCT.

Main Results

Unfortunately, KUR has been unavailable since May in 2014. Only 7 reports could be submitted.

RS1. Clinical studies on BNCT

Although no BNCT using KUR was performed, Fujimoto et al. reported a case report on BNCT for axillary lymph node metastasis of breast cancer which was carried out in 2013. In this report, BNCT successfully reduced the size of the lymph node metastases and alleviate the radiating pain in the arm. Case reports are very meaningful to consider the possibility of new applications of BNCT.

RS2. Pre-clinical studies on physiological and pharmacological aspects of BNCT

Yanagie et al. reported two pre-clinical studies. One was a preclinical BNCT study for VX-2 rabbit liver tumor model using borocaptate sodium (BSH) entrapped water-in-oil-in-water (WOW) consist with surfactant HCO40 or PGCR. Another one was a treatment planning study on BNCT for breast cancer

RS3. Medical physics studies on BNCT

Hayashi et al. investigated the influence of various lithium compounds of the dose response and the stability of polymer gel dosimeters.

Tanaka K et al. reported the imaging plate system to measure the beam components such as thermal, epithermal, fast neutrons and gamma rays separately.

Sakurai et al. studied the QA/QC in BNCT using ionization chamber and Bonner sphere in BNCT irradiation field.

Evaluation of Neutron Dosimetry for Boron Neutron Capture Therapy using Images of Breast Cancer

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INTRODUCTION: The JAERI Computational Dosimetry System (JCDS), which can estimate distributions of radiation doses in a patient's head by simulating in order to support the treatment planning for epithermal neutron beam BNCT, was developed. Kumada et al had reported that JCDS is a software that creates a 3-dimensional head model of a patient by using CT scan and MRI images, and that generates a input data file automatically calculation of neutron flux and gamma-ray dose distributions in the brain with the Monte Carlo code MCNP, and that displays these dose distributions on the head model for dosimetry by using the MCNP calculation results.

We evaluated the dosimetry of thermal neutron using JCDS in the condition of Single field irradiation or Bilateral irradiation to tumour constructed with MRI imagings of a breast cancer patient.

EXPERIMENTS: Neutron dosimetry with JCDS for a breast cancer patient: BNCT was simulated in a patient with a 7cm tumour in the lower half of the R breast. LiF collimation was used to selectively irradiate the tumor while sparing the adjacent normal organs (lung, heart). The Neutron Beam Facility at JRR4 enables to carry out boron neutron capture therapy with epithermal neutron beam.

RESULTS:

Simulation 1: Epithermal Neutron mode

Beam colimation : ϕ 7 c m, BPA : 24ppm, T/N=3.5, Restriction : Skin RBE dose as 10Gy-Eq

The blood concentration of BPA by drip infusion just before NCT was estimated as 24ppm, and T/N ratio was also estimated as 3.5 according to 18F-BPA-PET.

Simulation 1: Beam collimation was performed in

7cm field and epithermal neutron beam was irradiated with the tangenital direction (33min.) in restricted maximum skin RBE dose as 10Gy-Eq. The maximum tumor RBE dose was 87.5 Gy-Eq, the mean tumor RBE dose was 48.3 Gy-Eq, and the minimum tumor RBE dose was 11.5 Gy-Eq. The actual maximum skin RBE dose was 11.1 Gy-Eq (RBE=1.35).

Simulation 2: Epithermal Neutron mode

Beam colimation : φ 8 c m, BPA : 24ppm, T/N=3.5, Restriction : Skin RBE dose as 10Gy-Eq

Simulation 2: The beam direction was changed to oblique direction. The maximum tumor RBE dose was 86.6 Gy-Eq, the mean tumor RBE dose was 46.1 Gy-Eq, and the minimum tumor RBE dose was 19.3 Gy-Eq. The minimum tumor RBE dose was increased to 19.3 Gy-Eq from 11.5 Gy-Eq with the epithermal neutron in this oblique direction(70% up).

Simulation 3: Tangenital direction irradiation was performed in Simulation1, and Oblique direction irradiation was performed in Simulation 2. Combined two directional Irradiation was performed in Simulation 3 (70% of Simulation 1 (Irradiation time 23.9 min) + 79% of Simulation 2(Irradiation time : 23.9min)) in restricted maximum skin RBE dose as 10Gy-Eq.

The maximum tumor RBE dose was 107.3 Gy-Eq, the mean tumor RBE dose was 70.3 Gy-Eq, and the minimum tumor RBE dose was 28.0 Gy-Eq. This date shown that BNCT with two directional epithermal neutron irradiation can increase the RBE tumor dose to 2.4 times of Simulation 1 and 1.5 times of Simulation 2.

We hope to start BNCT clinical studies for recurrenced & advanced breast cancer patients. The irradiational directions will be decided using the JCDS or SERA simulation with the restriction of normal tissue tolerant RBE Dose.

We applied the JCDS to dosimetry of epithermal neutron, direction of neutron beam, and patient's positioning on BNCT. High resolution whole body dosimetry system, as JCDS and SERA will be very useful to evaluate the thermal/epithermal neutron dosimetry and the application of BNCT to recurring or advanced breast cancer.

Multi directional irradiations are hope to be available to increase the tumour RBE dose in the based on the tolerant dose of normal skin in BNCT field.

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Anti-tumor Effect of Boron Neutron Capture Therapy (BNCT) on Axillary Lymph NodeMetastasis of Breast Cancer

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INTRODUCTION: Breast cancer is the most morbid malignancy in women, and in Japan about 60,000 new cases are encountered every year. Although hormone therapy, chemotherapy and molecularly targeted therapy have improved its prognosis, 30% of such patients die from distant metastases. When systematic pharmacotherapy is not effective in metastatic cases, the disease is difficult to control. In this study, BNCT was assessed for lymph node metastasis of breast cancer (luminal type A). Additionally, the uptake of BPA was analyzed with the use of hormone-sensitive breast cancer cell lines.

PATIENT AND METHODS: (1) Patient: A 65-year-old woman diagnosed with left breast cancer (luminal A) and axillary lymph node metastasis underwent axillary lymph node dissection, mastectomy and radiation therapy for the area of the left axillary lymph nodes. Although subsequent lymph node and lung metastasis recurred, all tumors disappeared after both chemotherapy and hormone therapy. Nonetheless, local recurrence of tumors in the left lymph nodes was detected, for which chemotherapy was ineffective; therefore, intra-arterial chemo-embolization was carried out at a nearby clinic. The tumor was, however, not controlled; furthermore, palsy of the left axillary nerve was detected, and the patient complained of severe pain all over the upper left limb. Examination by Gd-enhanced MRI confirmed regrowth of the tumor; therefore, BNCT was administered.

(2) Boron concentration in tumor cell lines: Hormone-sensitive breast cancer cell lines (T47D, MCF-7), clear cell sarcoma (CCS) cell line (MP-CCS-SY) and melanoma cell line (G-361) were cultured and exposed to BPA (10, 20, 30 ¹⁰B μ g/ml) in the medium. Subsequently, the cells were washed, detached and collected; the concentration of ¹⁰B in the cells was then determined by ICP-AES.

RESULTS: An ¹⁸F-BPA-PET study conducted before BNCT showed accumulation of BPA in the tumor, with a

tumor to blood (T/B) ratio of 2.2. Since the tumor was located deep in the body, BNCT was administered by two-gate irradiation to the area of the left axilla, with the patient in a sitting position: ventrally, 5MW for 47min and dorsally, 5MW for 27min. The mean value of boron concentration in blood during BNCT was 29 ppm ventrally and 28.5 ppm dorsally, and neutron fluence on the surface of the body was 3.34×10^8 cm⁻²s⁻¹ ventrally and 4.89×10^8 cm⁻²s⁻¹ dorsally. The calculated average dose to the target and the left axillary nerve was 25 Gy-Eq and 4.3 Gy-Eq, respectively. Gd-enhanced MRI revealed that the recurrent tumor mass in the left axilla shrank two months after BNCT [Fig.1]. Moreover, the patient was free from the severe pain in the left arm, and the progress of paralysis ceased. Also, the two breast cancer cell lines demonstrated higher uptake of boron than did the CCS and melanoma cell lines that have already shown good outcome of BNCT [Fig. 2].

CONCLUSION: BNCT was effective in the treatment of metastatic breast cancer that had been resistant to all other methods of treatment. Additionally, in vitro studies demonstrated high uptake of BNCT by hormone-sensitive breast cancer cell lines. These data suggest the applicability of this new method of treatment to metastatic breast cancer.

FIGURES:







[Fig.2] Two hormone-sensitive breast cancer cell lines (T47D, MCF-7), CCS cell line (MP-CCS-SY) and melanoma cell line (G-361) were tested for their uptake of boron. Each cell line took up the boron in a concentration-dependent manner. The uptake of ¹⁰B by breast cancer cells was higher than that by the CCS and melanoma cells.

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PR8-3 Evaluation of Boron Concentrations in Liver by Intra-arterial Delivery of WOW Emulsion to VX-2 Hepatic Tumor Model for Neutron Capture Therapy to Hepatocellular Carcinoma

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INTRODUCTION: Water-in-oil-in-water(WOW) emulsion has been used as the carrier of anti-cancer agents by modifying of IPSO on intra-arterial injections in clinical. Higashi et al prepared a long term inseparable WOW for use in arterial injection therapy to treat patients with hepatocellular carcinoma(HCC) by the double emulcificating technique[1]. We performed preclinical BNCT study for VX-2 rabbit tumour model using ¹⁰BSH entrapped WOW [2, 3]. We also proceeded clinical BNCT study for HCC using this system[4].

In this study, we prepared ¹⁰BSH entrapped WOW in verifying the component of surfactant, and evaluated the boron delivery activity to measure the ¹⁰B concentrations of organs in VX-2 hepatic tumour model on time course after intra-arterial injection using ICP-Mas.

EXPERIMENTS: ¹⁰BSH entrapped WOW were administrated with intra-arterial injections via proper hepatic artery (¹⁰BSH : 20 mg/kg rabbit) on VX-2 rabbit hepatic tumour models. One and three days after arterial injections, the boron concentrations of the tumor nodules

and normal liver tissues were determinated by ICP- Mass Spectroscopy of Jyuntendo University.

RESULTS: VX-2-bearing rabbits (n = 3) were given intra-arterial injection with 2 ml of ¹⁰BSH WOW emulsion consist with surfactant HCO40, or PGCR. The mean ¹⁰B concentration prepared in ¹⁰BSH-WOW was 10000 ppm in this experiment. The size of WOW was controlled to 70 μ m.

The ¹⁰B concentration in VX-2 tumour was 170.8 ppm, 58.3 ppm by WOW with HCO40 after day1, day3 inta-arterial injection, respectively. The ¹⁰B concentration of tumour was 186.0ppm, 40.4ppm by WOW with PGCR after day1, day3 same injection, respectively. ¹⁰B concentration in normal liver tissue / blood were 8.0 / 0.3 ppm in HCO40 group, and 15.1 / 0.1 ppm in PGCR group at day 3, respectively in the same procedures of WOW.

We can deliver ¹⁰BSH to tumor site by intra-arterial injection with WOW emulsion. The effective ¹⁰B concentration (higher than 30ppm) was achieved in day1 and day3. It has be able to change the deliverable ¹⁰B atoms according to the sizes of WOW, and the type of surfactants. We hope to perform toxicity examinations of WOW emulsion to develop the more suitable WOW emulsion for intra-arterial born delivery system.

Table1. ¹⁰B concentration (ppm) of VX-2 hepatic tumor bearing rabbit model after intrarterial injection of ¹⁰B-WOW emulsion.

wow	Tumour	Normal liver		Blood
		Near Tumour	Another Lobe	
HCO40				
Day1	170.8 ± 27.3	31.9 ± 4.3	33.9 ± 30.9	0.73±0.4
Day3	58.3 ± 23.1	24.7 ± 17.1	8.02±1.7	0.33 ± 0.1
Day7	12.9 ± 7.5	2.9 ± 0.6	$1.7{\pm}0.4$	$0.12{\pm}0.0$
PGCR				
Day1	186.0±115.2	72.79 ± 2.5	69.1±42.1	1.27±0.5
Day3	40.47	56.7	15.1	0.14
Day7	40.66	37.3	13.8	0.14

The ¹⁰B conc. were determined by ICP-Mas. at Jyuntendo University.

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Establishment of QA/QC Using Ionization Chamber and Bonner Sphere in BNCT Field (II)

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INTRODUCTION: After the restart of the operation of Kyoto University Reactor (KUR) in May 2010, 235 clinical studies of boron neutron capture therapy (BNCT) have already been carried out as of May 2016 [1]. Also, the BNCT clinical trial using Cyclotron-based BNCT Epi-thermal Neutron Source (C-BENS) started in November 2012 [2]. In the while, the research and development into several types of accelerator-based irradiation systems are underway by several research groups in the world at present time. With this situation in mind, it is important that the physical and biological estimations for dose quantity and quality are performed consistently among several irradiation fields, and that the equivalency of BNCT is guaranteed, even across BNCT systems. The aim of this research is the establishment of quality assurance and quality control (QA/QC) in BNCT irradiation field. As part of the QA/QC system, we are developing estimation method for neutron energy spectrum using Bonner sphere.

METHODS: For our spectrometer using Bonner sphere, liquid such as pure water and/or boric acid solution is used as the moderator [3]. As shown in Fig. 1, a multi-layer concentric-sphere case with several sphere shells is prepared. The moderator and its diameter are changeable without entering the irradiation room, by the remote supply and drainage of liquid moderator in the several layers. For the detector, activation foils are remotely changed, or online measurement is performed using SOF (scintillator with optical fiber) detector containing boron, etc.. On the assumption of the application in a typical BNCT irradiation field, the combination of the moderators for boron-10 (B-10) concentration and diameter was optimized by our originally-developed method, "High Independence Selection (HIS)" [4]. For the B-10 concentration, the selection was performed among ten values such as 0, 0.01, 0.016, 0.028, 0.048, 0.082, 0.14, 0.24, 0.41 and 0.7 weight percent (wt%). For the diameter, the selection was performed among ten values from 11 to 20 cm in 1 cm increment. Manganin foil was assumed to be used as the detector, which has high response mainly to thermal neutrons. The optimized combination was decided among one hundred and one combinations; the combinations of ten B-10 concentrations and ten diameters, additionally the case of manganin foil only without the moderator.

RESULTS: The optimized combination was selected by HIS as follows: manganin foil only, 0.7-wt% boron acid solution of 13 cm in diameter, 0.7-wt% boron acid solution of 18 cm in diameter, 0-wt% boron acid solution (namely pure water) of 18 cm in diameter, and

0.028-wt% boron acid solution of 20 cm in diameter. Then, the optimized structure of the spectrometer was decided as follows: three sphere shells such as 13, 18 and 20 cm in diameter, and three liquid moderators such as pure water, 0.028-wt% boron acid solution and 0.7-wt% boron acid solution. It is not thought that this structure is necessary to be changed when the detector is changed from manganin foil to boron-containing SOF detector.

CONCLUSION: We have a plan to make the Bonner-sphere spectrometer, based on the optimization result. Additionally, we have a plan to perform the spectrometry experiments at Kyoto University Reactor (KUR), etc., in order to confirm the efficacy of this spectrometer.



Fig. 1. Concept design of the Bonner-sphere spectrometer for QA/QC in BNCT.

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PR8-5 A Consideration of the Influence by the Radiation Type on the Measurement of the Spatial Distributions of the Beam Components in BNCT Using the Imaging Plate

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INTRODUCTION: For boron neutron capture therapy (BNCT), measurement of the spatial distributions of neutrons and gamma rays is one of the potential options for the quality assurance and quality control. It is desirable to measure the beam components such as thermal, epithermal, fast neutrons and gamma rays, separately. This study investigates the usage of the imaging plate (IP) for this purpose. The influence by the radiation type which deposits the energy in the IP on the resultant beam component distribution is discussed here.

METHODS: Two IPs set in a converter were irradiated^[1] at the collimator aperture of the standard epithermal neutron irradiation mode at the Kyoto University Reactor Heavy Water Neutron Irradiation Facility (KUR-HWNIF). The converters were the epoxy resin doped with boron which is attempted to enhance epithermal neutrons via secondary particles from the ¹⁰B(n, α)⁷Li reaction, and carbon which is not expected to enhance neutrons due to its low neutron interaction. The fluence ϕ was determined using the following model;

$$PSL = \begin{pmatrix} PSL_1 \\ PSL_2 \end{pmatrix} = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \begin{pmatrix} \phi_1 \\ \phi_2 \end{pmatrix} = A \cdot \phi$$
(1),
$$\phi = A^{-1} \cdot PSL$$
(2),

where PSL_i denotes the signal intensity of the *i*th IP, a_{ij} the sensitivity of the *i*th IP for the component *j*. For *j*, 1 denotes gamma rays, and 2 epithermal neutrons. In this analysis, the energy deposition calculated by the PHITS code was used as a_{ij} . The problem is that the dependence of the radiation type on the efficiency that the energy deposition is converted to the IP signal is not included. In the present report, the dependence of the IP signal on the radiation type is investigated from the previous studies. Then, its influence on the experimental result of the component distributions is discussed.

RESULTS and DISCUSSION: The dependence of the IP signal intensity on the radiation type that deposits the energy in the sensitive region of the IP, or its LET, has been investigated during past years. However, the mechanism or its quantitative influence has not been concluded consistently among the publications. However, there are several proposals on it.

One of the earliest and most comprehensive studies was conducted by Thoms in 1996^[2]. They categorized the processes potential to be influenced into two, i.e., readout of the IP signal, and fading of the signal.

As to the readout, the influential process is considered to be the attenuation of the readout light emitted from the sensitive region, the efficiency of converting the energy deposition into the IP signal will be higher for the alpha particles than that for the gamma rays. Bennet [3] reported that the readout light is attenuated to 1/e (37%) by the IP sensitive region with the thickness of 113-211 µm the IP by Fujifilm Co. Ltd used in the experiment^[1]. The sensitive region has the thickness of 50 µm and it is supposed to attenuate the readout light by 21-36 %. In addition, the readout light spreads and results in decrease of the IP signal intensity. The decrease was reported to be by 13 % for the thickness of 50 μ m by Thoms^[2]. Alpha particles and ⁷Li nuclei deposits its energy within a few micro meters from the IP surface, while the recoil protons and secondary electrons from the converters deposit the energy in whole the thickness of 50 µm. The efficiency of the energy deposition to be the IP signal will be higher for the former, by possibly a few tens percents. Accordingly, the contribution of the epithermal neutron components to the IP signal is expected to be slightly higher than the calculated sensitivities for the equation (1).

On the other hand, the time dependence of the fading of the IP signal has been reported to be independent from the radiation type^[2,4]. However, the results of investigation by Nakamura^[5] showed that the IP signal produced by alpha particles reduces more rapidly than those by X-rays. At 30 minutes or more after the irradiation, which corresponds to the experiment^[1], the difference was reported to be about 30 to 40%.

These factors, i.e., the readout light attenuation and IP signal fading, have opposite influences, i.e., the former increases the sensitivity to the alpha particles, and the latter decreases. In total, the type of radiation could double the IP signal, while it could be an overestimation. To include its influence, the sensitivity a_{ii} should be adjusted. For example, if the sensitivity for the secondary particles of ${}^{10}B(n,\alpha)^7Li$ reaction that enhances the epithermal neutron component is β times, a_{i2} for all the IPs should be replaced with βa_{i2} . Consequently, the solution will be $\phi_{epi}' = \phi_{epi}/\beta$. This means that including the influence by the radiation type does not change the relative distribution of the resultant fluences, which is what this study aimed to assure. The analysis without considering the dependence of IP signal on radiation type will be enough to assure the temporal change in the spatial distribution of the irradiation field.

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PR8-6

Development and Evaluation of 3D Polymer Gel Dosimeter for the Measurement of dose Distribution in BNCT

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INTRODUCTION: Polymer gel dosimeters have been investigated for the three-dimensional (3D) dose measurement of the complex conformal dose distributions in the clinical applications [1]. These devices utilize radiation-induced polymerization reaction of vinyl monomer in the aqueous gel matrix to preserve information about the radiation dose. The 3D absorbed dose distribution is deduced from the created polymer distribution measured by imaging modalities such as MRI and Optical CT.

Polymer gel dosimeter is also regarded as tissue equivalent to neutron beam because the components are mainly water and a small amount of other chemicals consisting of carbon, nitrogen and oxygen. A further advantage of polymer gel dosimeters is that the interaction with neutron could be controlled by addition of some compounds with neutron-capture-nuclei such as ⁶Li and ¹⁰B. It means that each dose component might be distinguished from complex dose due to various primary and secondary radiations by the variety of elemental composition.

In the previous work, we have investigated that the dose response of polymer gel dosimeter irradiated by thermal neutron beam was enhanced by addition of a very small amount of ¹⁰B (50 ppm) [2]. In this work, the influence of various lithium compounds on the dose response and the stability of polymer gel dosimeters was investigated by high-energy photon beam before the future experiments using neutron beam.

EXPERIMENTS: In this work, MAGAT-type (methacrylic-acid-based) polymer gel dosimeter was employed because of the high sensitivity. Various lithium salts (LiCl, LiNO₃, LiOH, Li_2SO_4 and Li_3 -citrate, which have comparatively high solubility in water) containing ⁶Li in the natural ratio were mixed into the polymer gel in the concentration of

100 and 200 ppm of ⁶Li, respectively. The resulting gels were subdivided by pouring into test tubes.

The irradiations were performed using 6 MV X-ray from a medical linear accelerator (No-varis-TX, Varian/BrainLAB) and gamma ray from ⁶⁰Co source. The doses upto 5 Gy were delivered to each sample.

The read-out from the samples was performed using a 1.5 T MRI scanner (SIGNA HDxt 1.5T, GE Healthcare) with a head coil the day after irradiation. A multiple spin-echo sequence was applied and the transverse relaxation rate R_2 (=1/ T_2) was obtained as the function of absorbed dose.

RESULTS: In the results, the dose- R_2 responses (sensitivities) of the polymer gel dosimeters containing LiNO₃, LiOH and Li₃-citrate decreased significantly (40-70% lower at 5 Gy) comparing with that of the basic gel dosimeter without lithium salts. Also, Li₃-citrate did not dissolve enough in the gel solution and the gel was translucent. On the other hand, the gel dosimeters containing LiCl and Li₂SO₄ showed the comparable sensitivity to the basic gel dosimeter and the rather sensitivity enhancement was also observed.

From these results, it was suggested that LiCl or Li_2SO_4 was a suitable additive to introduce lithium to the polymer gel dosimeter. When lithium salts containing the enriched ⁶Li could be used, the clearer enhancement due to ⁶Li would be expected even in a smaller amount of additive.

(These results would be presented at 17th International Congress on Neutron Capture Therapy, ICNCT-17 in Missouri, USA.)

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PR8-7 Patient-Position-Error Measuring System for BNCT Irradiation

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INTRODUCTION: In boron neutron capture therapy (BNCT) irradiations carried out at Kyoto University Research Reactor, sitting position has been applied in many cases, considering a flexibility of patient positioning and structural restriction of an irradiation facility. In those cases, the patient position is sometimes unstable, resulting in a displacement from an initial set-up position determined by a treatment planning process. The displacement and motion during an irradiation period cause uncertainty in estimation of delivered dose.

Aiming to improve the dose estimation accuracy, we have prepared a patient-position-error measuring system using a motion sensor. An outline of the measuring system and initial test operation are described.

MATERIALS AND METHODS: An outline of the measuring system is shown in Fig. 1. A MEMS motion sensor module (IMU-Z 2, ZMP Inc., Tokyo) including tri-axial accelerometer, gyroscope and magnetometer was used to track a position and a rotation angle during an irradiation period. The sensor module $(36 \times 52 \times 11 \text{ mm})$ and a battery box $(36 \times 62 \times 15 \text{ mm})$ are sufficiently small. In addition, the module has a wireless data transmission function. These features enable us to fix the sensor on patient surface without interference in patient setup and irradiation process.

A displacement can be calculated by integrating accelerometer data twice with respect to time. Also, a rotation angle can be calculated by integrating gyroscope data with respect to time in motion phase and by direction of gravitational acceleration and geomagnetic field at resting phase. The estimated displacement and rotation angle are displayed and saved as a log data over an irradiation period. The log data can be utilized for post-irradiation dose evaluation.

This system is considered to be useful for head cases, where the measuring object is regarded as a rigid body. For those cases such as brain tumor or head and neck tumor, the sensor module and battery box are fixed on top of head so that the sensor coordinate system is aligned to anterior-to-posterior, left-to-right and inferior-to-superior directions of the patient. The position and rotation angle of the head is measured in the reference coordinate system defined on the irradiation room.

RESULTS: An initial test operation of the motion sensor was conducted. Time fluctuations were observed in the accelerometer output as well as the gyroscope and magnetometer. High frequency component of the accelerometer output, related to a sensor noise, was observed in the amplitude range of about 0.3% with respect to the gravitational acceleration. This component can be removed by using an appropriate high-pass filter. Low fre-

quency component, related to a sensor bias, showed time-dependent behavior, which resulted in abnormal divergence of velocity obtained from integration of raw accelerometer output. This phenomenon is well known as velocity drift in general applications of motion sensor. The velocity drift can be corrected at a resting state, at which velocity is adjusted to zero. The resting state need to be detected based on the sensor outputs using appropriate threshold values. These filtration and correction are under examination and need to be optimized to clinical situation of BNCT irradiation.



Fig. 1. An outline of patient-position-error measuring system.

PR9 Analyzing Tumor Microenvironment and Exploiting its Characteristics in Search of Optimizing Cancer Therapy Including Neutron Capture Therapy

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BACKGROUNDS AND PURPOSES: Human solid tumors contain moderately large fractions of quiescent (Q) tumor cells that are out of the cell cycle and stop cell division, but are viable compared with established experimental animal tumor cell lines. The presence of Q cells is probably due, in part, to hypoxia and the depletion of nutrition in the tumor core, which is another consequence of poor vascular supply. As a result, Q cells are viable and clonogenic, but stop cell division. In general, radiation and many DNA-damaging chemotherapeutic agents kill proliferating (P) tumor cells more efficiently than O tumor cells, resulting in many clonogenic O cells remaining following radiotherapy or chemotherapy. Therefore, it is harder to control Q tumor cells than to control P tumor cells, and many post-radiotherapy recurrent tumors result partly from the regrowth of Q tumor cells that could not be killed by radiotherapy. Similarly, sufficient doses of drugs cannot be distributed into Q tumor cells mainly due to heterogeneous and poor vascularity within solid tumors. Thus, one of the major causes of post-chemotherapy recurrent tumors is an insufficient dose distribution into the Q cell fractions.

With regard to boron neutron capture therapy (BNCT), with ¹⁰B-compounds, boronophenylalanine-¹⁰B (BPA) increased the sensitivity of the total cells to a greater extent than mercaptoundecahydrododecaborate-¹⁰B (BSH). However, the sensitivity of Q cells treated with BPA was lower than that in BSH-treated Q cells. The difference in the sensitivity between the total and Q cells was greater with ¹⁰B-compounds, especially with BPA. These findings concerning the difference in sensitivity, including other recovery and reoxygenation following neutron irradiation after ¹⁰B-compound administration were mainly based on the fact that it is difficult to deliver a therapeutic amount of ¹⁰B from ¹⁰B-carriers throughout the target tumors, especially into intratumor hypoxic cells with low uptake capacities.

Hypoxia is suggested to enhance metastasis by increasing genetic instability. Acute, but not chronic, hypoxia was reported to increase the number of macroscopic metastases in mouse lungs. We recently reported the significance of the injection of an acute hypoxia-releasing agent, nicotinamide, into tumor-bearing mice as a combined treatment with y-ray irradiation in terms of repressing lung metastasis. As the delivered total dose increased with irradiation, the number of macroscopic lung metastases decreased reflecting the decrease in the number of clonogenically viable tumor cells in the primary tumor. The metastasis-repressing effect achieved through a reduction in the number of clonogenic tumor cells by irradiation is much greater than that achieved by releasing tumor cells from acute hypoxia. On the other hand, more ¹⁰B from BPA than from BSH could be distributed into the acute hypoxia-rich total tumor cell population, resulting in a greater decrease in the number of highly clonogenic P tumor cells with BPA-BNCT than with BSH-BNCT and with neutron beam irradiation only.

BPA-BNCT rather than BSH-BNCT has some potential to decrease the number of lung metastases, and an acute hypoxia-releasing treatment such as the administration of nicotinamide or bevacizumab may be promising for reducing numbers of lung metastases. Consequently, BPA-BNCT in combination with nicotinamide and/or bevacizumab treatment may show a little more potential to reduce the number of metastases. Now, it has been elucidated that control of the chronic hypoxia-rich Q cell population in the primary solid tumor has the potential to impact the control of local tumors as a whole, and that control of the acute hypoxia-rich total tumor cell population in the primary solid tumor has the potential to impact the control of lung metastases.

The aim of this research project is focused on clarifying and analyzing the characteristics of intratumor microenvironment including hypoxia within malignant solid tumors and optimizing cancer therapeutic modalities, especially radiotherapy including BNCT in the use of newly-developed ¹⁰B-carriers based on the revealed findings on intratumor microenvironmental characteristics.

RESEARCH SUBJECTS:

- The collaborators and allotted research subjects (ARS) were organized as follows;
- **ARS-1 (27P9-1)**: Optimization of Radiation Therapy Including BNCT in terms of the Effect on a Specific Cell Fraction within a Solid Tumor and the Suppressing Effect of Distant Metastasis. (<u>S. Masunaga, et al.</u>)
- ARS-2 (27P9-2): Development of Hypoxic Microenvironment-Oriented ¹⁰B-Carriers. (<u>H. Nagasawa</u>, *et al.*)
- **ARS-3** (27P9-3): Clarification of Mechanism of Radio-Resistance in Cancer Using Optical Imaging at Tissue Level. (<u>H. Harada</u>, *et al.*)
- **ARS-4** (27P9-4)*: Analysis of Radiation-Induced Cell-Killing Effect in Neutron Capture Reaction. (<u>R.</u><u>Hirayama</u>, *et al.*)
- **ARS-5** (27P9-5)*: Transdermal Drug Delivery System using Hyaluronan-Conjugated Liposomes as ¹⁰B-Carrier in Boron Neutron Capture Therapy for Melanoma (<u>S. Kasaoka, et al.</u>)
- **ARS-6** (27P9-6) *: Evaluation of Inclusion Complex of Carborane Modified Kojic Acid and Cyclodextrin as ¹⁰B-Carrier in Boron Neutron Capture Therapy. (<u>T. Na-gasaki</u>, *et al.*)
- **ARS-7** (27P9-7)*: Molecular Design and Synthesis and Functional Evaluation of Anticancer and Molecular Targeting Agents. (<u>Y. Uto</u>, *et al.*)
- **ARS-8** (27P9-8)*: Analyzing Biological Effect of BNCT from the Viewpoint of the Changes in Oxygenation Level. (<u>H. Yasui</u>, *et al.*)
- ARS-9 (27P9-9): Analyses on the Responsiveness of Malignant Tumors to BNCT. (<u>M. Masutani</u>, *et al.*)
- **ARS-10 (27P9-10)** *: Assay for Tumor Cell Survival and Tumor Growth Delay through Neutron Capture Reaction according to the Changes in Intracellular Concentrations within Solid Tumors of Newly- Developed ¹⁰B-Carriers. (K. Nakai, *et al.*)
- **ARS-11** (27P9-11)*: Antitumor and Metastasis-Reppresing Effect of BNCT on Human Breast and Pancreatic Cancer Cell Lines. (<u>Y. Matsumoto</u>, *et al.*)
- (* Due to the absence of operating our reactor in 2015, no data were obtained, resulting in no reporting here.)

PR9-1 The Effect of *p53 Status* of Tumor Cells on Radiosensitivity of Irradiated Solid Tumors with Accelerated Carbon-ions Compared with γ-rays or Reactor Neutrons

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INTRODUCTION: We examined the characteristics of radio-sensitivity in the total (proliferating (P) plus quiescent) and quiescent (Q) cell populations in solid tumors irradiated with 290 MeV/u accelerated carbon ion beams at varying LET values in a 6-cm spread-out Bragg peak (SOBP) installed at the National Institute of Radiological Sciences (Chiba, Japan) compared with irradiation with ⁶⁰C γ -rays and reactor thermal and epithermal neutron beams at our institute with our method for selectively detecting the response of Q cells within solid tumors [1,2], using two different tumor cell lines with identical genetic backgrounds except for *p53* status.

MATERIALS AND METHODS: Human head and neck squamous cell carcinoma cells transfected with mutant TP53 (SAS/mp53) or with neo vector (SAS/neo) were injected subcutaneously into hind legs of nude mice. Tumor-bearing mice received 5-bromo-2'-deoxyuridine (BrdU) continuously to label all intratumor P cells. They received γ -rays or accelerated carbon-ion beams at a high or reduced dose-rate. Other tumor-bearing mice received reactor thermal or epithermal neutrons at a reduced dose-rate. Immediately or 9 hours after the high dose-rate irradiation (HDRI), or immediately after the reduced dose-rate irradiation (RDRI), the tumor cells were isolated and incubated with a cytokinesis blocker, and the micronucleus (MN) frequency in cells without BrdU labeling (= Q cells) was determined using immunofluorescence staining for BrdU.

RESULTS: The difference in radio-sensitivity between the total (= P + Q) and Q cells after γ -ray irradiation was markedly reduced with reactor neutron beams or carbon ion beams, especially with a higher linear energy transfer (LET) value. Following γ -ray irradiation, SAS/*neo* tumor cells, especially intratumor Q cells, showed a marked reduction in sensitivity due to the recovery from radiation-induced damage, compared with the total or Q cells within SAS/*mp53* tumors that showed little repair capacity [3]. In both total and Q cells within both SAS/*neo* and SAS/*mp53* tumors, carbon-ion beam irradiation, especially with a higher LET, showed little recovery capacity through leaving an interval between HDRI and the assay or decreasing the dose-rate. The recovery from radiation-induced damage after γ -ray irradiation was a p53-dependent event, but little recovery was found after carbon-ion beam irradiation. With RDRI, the radiosensitivity to reactor thermal and epithermal neutron beams was slightly higher than that to carbon ion beams.

DISCUSSION: Two major pathways for the repair of potentially lethal DNA dsbs exist in mammalian cells. The non-homologous end-joining (NHEJ) pathway is imprecise, error-prone, and mutagenic, and mutant cell lines lacking key components of this pathway all exhibit impaired kinetics of DNA dsb repair and exquisite radio-sensitivity. Homologous recombination (HR) is a more precise (error-free) repair mechanism and is more important for the repair of dsbs in late-S and G2 when a sister chromatid is available for the recombination reaction. Cell lines with defects in HR also exhibit increased radio-sensitivity and decreased fidelity of repair.

A cellular safeguard against genetic destabilization is activation of the *p53* tumor suppressor protein, which commonly responds to DNA damage signals by inducing apoptosis or regulating the cell cycle including DNA damage repair. As also shown in our previous report, the net MN frequencies in SAS/*neo* tumor cells were lower than those in SAS/*mp53* tumor cells under all conditions (P < 0.05), probably resulting from exclusion of a higher number of radiation-induced apoptotic SAS/*neo* cells than SAS/*mp53* cells.

Loss-of-function of *wild-type TP53* can result in loss of the G1/S cell-cycle checkpoint and an increase in HR. As *p53* seems to interact with RAD51, the absence of normal *p53* function is thought to enhance RAD51-mediated DNA pairing activity and HR, due to overexpression of RAD51 out of regulation by normal *p53*. Thus, HR is thought to be a principal mechanism of DNA dsb repair in SAS/*mp53* cells. The very small repair capacity of SAS/*mp53* cells *in vivo* may show that the repair in solid tumors with a mutant *p53* is mainly due to, if anything, the NHEJ rather than HR.

CONCLUSION: For tumor control, including intratumor Q-cell control, accelerated carbon-ion beams, especially with a higher LET, and reactor thermal and epithermal neutron beams were very useful for suppressing the recovery from radiation-induced damage irrespective of p53 status of tumor cells [4].

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PR9-2 Development of New Membrane Penetrating Boron Carriers Comprised of BSH and Pepducins

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INTRODUCTION: Selective delivery of sufficient quantity of 10B to tumor cells is essential for the success of boron neutron capture therapy (BNCT). Although sodium borocaptate (BSH) has been used clinically as a boron carrier for BNCT, it is impermeable to plasma membrane due to its high hydrophilicity and anionic charges. Recently, we found that pepducin, which are artificial lipopeptides derived from an inner loop domain of G protein-coupled receptors (GPCRs), ¹ enabled anionic molecule such as fluorescein to penetrate membrane directly.² These findings let us to envisage that an anionic boron cluster can be delivered into cytosol using the pepducin delivery unit.

In this study, we designed and synthesized novel pepducin-boron cluster hybrid compounds as boron carriers for BNCT and evaluated the intracellular delivery of them.

MOLECULAR DESIGN: As shown in Fig. 1, we designed new hybrid molecules comprising pepducin (Pep) as a vehicle, and boron cluster (BS) as a cargo through an appropriate linker. When this molecule is internalized into the cytosol, intracellular glutathione can cleave the disulfide bond (SS) to release the boron cluster cargo into the cytosol. 10B atoms are accumulated in the cells due to hydrophilicity of the anionic property.



Fig. 1 Design of membrane penetrating boron carriers

METHODS: To explore the structural requirements of membrane penetrating boron carrier for intracellular uptake, we evaluated the sequence and length of peptide and structure of lipid moiety. Various lipidated peptides were prepared by solid-phase synthesis and then combined with BSH through an appropriate liker to afford the boron carrier. T98G cells were treated with the boron carriers (10 μ M) at 37 °C for various time, then, washed with PBS three times, and dissolved in 200 μ L HNO₃ for 1 h. The boron concentrations of the extracts were measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES: HORIBA JOVIN YBON UL-TIMA2) detected at 249.773 nm.

RESULTS AND DISCUSSION: Various lipopeptide -BSH hybrids were synthesized (Table 1). Compound 13Pep showed the highest boron concentration in cells

Table 1 Structures of boron carriers



among the test compounds examined by ICP-AES. The boron concentration increased over time from 2h to 12 h. (Fig. 2). These data suggested that the palmitoyl tail and

AVANRSKKSRALF

AVANRSKKSR

 CH_3

C15H31

Ac13pep

10Pep





hydrophobic residues at C-terminal end are necessary for the intracellular penetration.

We are performing the further optimization of peptides and linker structures to promote the intracellular accumulation.

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PR9-3 LY6E; a Conductor of Malignant Tumor Growth through Modulation of the PTEN/PI3K/Akt/HIF-1 Axis

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INTRODUCTION: Accumulating evidence has shown that hypoxia-inducible factor 1 (HIF-1) plays critical roles in radioresistance of cancer cells and tumor recurrence after radiation therapy and eventually causes death among cancer patients. Clinical studies have demonstrated consistent data that HIF-1 could be used as an adverse prognostic factor for not only local tumor recurrence but also distant tumor metastasis in cancer patients. These findings justify targeting HIF-1 for cancer therapies [1,2].

HIF-1, a heterodimeric transcription factor com-posed of an α -subunit (HIF-1 α) and a β -subunit (HIF-1 α), its activity is known to depend on the expression levels of HIF-1a protein. Under normoxic conditions, HIF-1a protein is actively degraded through the hydroxylationand subsequent ubiquitination-mediated proteolysis reactions. On the contrary, HIF-1a becomes active under hypoxic conditions because of the inactivation of the hydroxylases, and then, interacts with its binding partner, HIF-1β. Resultant heterodimer, HIF-1, binds to its cognate enhancer sequence, the hypoxia-responsive element (HRE), and induces transcriptions of various genes related to the escape from hypoxia (invasion and metastasis of cancer cells) as well as the improvement of oxy-gen-availability (angiogenesis) and adaptation of cellular metabolism to hypoxia (metabolic reprogramming).

In order to explore novel genes which are responsible for the HIF-1-mediated tumor malignant progression, we recently established a new genetic screening method and found that overexpression of lymphocyte antigen 6 complex, locus E (LY6E) is responsible for the activation of HIF-1. In the present study, we analyzed both the molecular mechanisms underlying the LY6E-mediated activation of HIF-1 and the involvement of LY6E-HIF-1 axis in malignant progression of cancers.





EXPERIMENTS & RESULTS: Forced expression of LY6E using a plasmid-based expression vector for LY6E increased HIF-1 α gene expression principally at the transcription level (Fig. 1).

This, in turn, led to the expression of the pro-angiogenic factors, VEGFA and PDGFB, through decreases in the expression levels of PTEN mRNA and subsequent activation of the PI3K/Akt pathway. The LY6E-HIF-1 axis functioned to increase tumor blood vessel density and promoted tumor growth in immune-deficient mice (Fig. 2).





LY6E expression levels were significantly higher in human breast cancers than in normal breast tissues, and were strongly associated with the poor prognoses of various cancer patients (Fig. 3). Moreover we found that LY6E induced radioresistance of cancer cells via the activation of HIF-1.





Our results characterized LY6E as a novel conductor of tumor growth and tumor radioresistance through its modulation of the PTEN/PI3K/Akt/HIF-1 axis and demonstrated the validity of targeting this pathway for cancer therapy.

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INTRODUCTION: Boron neutron capture reaction (BNCR) efficiently introduces DNA double strand breaks in the cells [2], however, tumor cell killing is affected by various factors [3] including the uptake of boron compounds. Heterogeneiety of cancer cells in the tumor tissues, such as the presence of cancer stem-like cells and hypoxic cells, may potentially cause resistant populations to the boron neutron capture therapy (BNCT) [1]. It is therefore important to analyze the responses of the cells to boron neutron capture reaction (BNCR) in various conditions. We have performed comprehensive analysis of mRNA expressions and proteome of human squamous carcinoma SAS cells after BNCR [4]. Changes in the protein levels involved in the various functions, such as endoplasmic reticulum, DNA repair, and RNA processing were observed within 24 hrs after neutron-beam irradiation. We also showed that BNCR induced generation of fragments from endoplasmic reticulum-localized lymphoid-restricted protein (LRMP). The elucidation of biological significance of fragmentation of LRMP is further necessary.

EXPERIMENTS: Because the neutron-beam irradiation with KUR nuclear reactor was not carried out during FY2015, we analyzed the irradiated cell samples prepared before. We used the cell extract of human oral squamous cancer SAS cells [1] to perform the mRNA expression and proteome analysis. Previously, ¹⁰B-boronophenylalanine (¹⁰B-BPA)-fructose solution was prepared as described [5]. SAS cells were suspended in the polypropyrene vials and incubated 2 hrs with or without 25 ppm of ¹⁰B-BPA [5]. After 6 and 24 hrs irradiation operated at 1 MW in the KUR facility, cells were separated to supernatant and cell pellet. Proteome analysis was performed using two dimensional polyacrylamide gel electrophoresis and mass spectrometry. Thermal neutron fluence and gamma-ray dose was measured with thermaluminescence dosimeter. These physical radiation doses were measured with the kind help of Drs. Yoshinori Sakurai and Hiroki Tanaka of KUR. ¹⁰B concentration was measured by prompt-gamma ray analysis (PGA) as described elsewhere.

RESULTS: For the analysis of RNA and protein dynamics, we used therapeutic dose conditions. The irradiated total dose of BNCR-treated SAS cells was about 17.5 Gy and that of neutron beam-irradiated SAS cells was about 4.0 Gy. Cell lysates of SAS cells with BPA (BNCR-sample) or without BPA (control) harvested 6 and 24 hrs post-irradiation were analyzed. Twenty-four hours after irradiation, apoptosis was observed as a major cell death and cleavage of caspase-9, caspase-3 and PARP-1 was observed. The sign of necrosis was also observed at this time point. The expression analysis of mRNA demonstrated dynamic changes of various genes related to inflammation and immune responses, and transcription. In the proteomic analysis [4], the peptide sequences from twenty-two spots have been determined by MALDI-TOF/MS from the twenty-nine spots that showed changes in the intensities between BNCR-sample and control.

We observed that proteins involved in the vesicle regulation, mRNA processing, transcription, rRNA processing, GTPase activity, ribosome biogenesis, DNA replication, and respiratory electron transport chain showed dynamic changes. Notably, lymphoid-restricted membrane protein (LRMP) and steroid hormone receptor (ERR1) were detected in multiple spots of different molecular mass. LRMP/ Jaw1 is known to be present at the cytoplasmic face of the ER, but its function has not been well understood [6]. We observed fragmentation of LRMP in the grafted lymphosarcoma of the rat 20 hrs after BNCR [4].

We further analyzed the LRMP in BNCR-sample and control by western blotting. Truncated LRMP of around 22 kDa was induced 24 hrs after BNCR-irradiation. The study for the biological significance of truncated LRMP in BNCR-induced cell death is ongoing.

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Production Mechanism of Radioactive Aerosols Released from Fukushima Daiichi Nuclear Power Plant

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OBJECTIVE: Environmental distribution and transition of radioactive materials released from Fukushima Daiichi Nuclear Power Plant has been investigated by means of air dose rate measurements and radioactivity measurements for environmental samples. On the other hand, chemical forms of the radioactive materials released from the power plant and transition mechanisms in the environment has not been revealed. The chemical form of the radioactive materials just behind the release is important to evaluate the internal exposure by radioactive aerosols and the transition of the materials in soils and plants after the fall out. A radioactive aerosol is one of the most important transport medium of the radioactive materials. In the present project research, the production mechanism of radioactive aerosols has been studied to elucidate the chemical form of the radioactive materials released from the power plant.

RESEARCH SUBJECTS: The project research is composed by four individual subjects in this year. Two are developments of production method of radioactive aerosols for simulation experiments, one is development of measurement method of radioactive aerosols, and one is elucidation of the production mechanism of aerosol under the specific situation. The respective subjects of the research groups of this project are described as follows;

P10-1: Development of production method of radioactive aerosols by attaching radioisotopes to aerosol particles

P10-2: Development of measurement method of radioactive aerosols under severity conditions

P10-3: Development of production method of radioactive aerosols by heating radioisotopes

P10-4: Study for production mechanism of radioactive aerosols in the presence of sea water

In this year period, experiments of subjects P10-3 and P10-4 were not carried out because of no operation of KUR.

RESULTS: In the subject P10-1, an attachment ratio of fission products to aerosol particles was estimated using a solution aerosol production apparatus and a spontaneous fission source of ²⁵²Cf in order to elucidate the attachment mechanism of fission products to aerosol particles.

The solution aerosols were generated from alkali halide (NaCl, NaBr, NaI, CsCl) and ammonium sulfate solution of 0.1 wt% concentration using an atomizer. The attachment ratio can be considered by associating with surface area of aerosol particles definitely because aerosols classified by size were used in the experiments. The relationships between the surface area and the estimated attachment ratio for a fission product of ¹⁰⁴Tc were fitted by a function on the basis of the assumption of the adsorption-desorption equilibrium between fission products and surface of aerosol particles. From results of the fitting, equilibrium constants and saturated attachment ratios were obtained for the combination of ¹⁰⁴Tc and each solution aerosol. In the comparison among the estimated equilibrium constants of ¹⁰⁴Tc for solution aerosols, it was found that the equilibrium constant for sodium chloride solution aerosol is smaller than those for other solution aerosols. This difference might be caused by density profiles of the anions in the fine solution particles that is explained by molecular dynamics simulations.

For the development of measurement method of radioactive aerosols under severity conditions, an air irradiation experiment was performed using the FFAG (Fixed Field Alternating Gradient) proton accelerator, and a combination method of imaging plate (IP) measurement and the wire screen technique was tried to estimate particle size of ¹¹C-bearing aerosols which were produced in the air irradiation. The ¹¹C activities collected by each screen and the backup filter were estimated from the decay analysis. The activity ratio of particles penetrating the screens to whole particles before penetrating the screens is governed by their diffusion coefficient expressed by a function of particle size. The size distribution was calculated by fitting the curve of the penetrating ratios vs. the number of screens to the theoretical equation. Log-normal distributions are assumed as the size distribution function in the calculation because the number-based size distribution was confirmed to be log-normal by another measurement method. The geometric mean diameter for the ¹¹C-bearing aerosol particles was successfully obtained using the combination method of the wire screen collection and the IP measurement. The activity-based geometric mean diameters were larger than the number-based one by ca. 20 nm. This shows that attachment process of radioactive atoms to the surface of aerosol particles is included in the radioactive particle formation.

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INTRODUCTION: Almost radioactive materials released in the air from Fukushima Daiichi Nuclear Power Plant have been transported by aerosols. Measurement results about size distributions and activity size distributions of the radioactive aerosols have been reported, and it was suggested that sulfate compounds were the potential transport medium for radioactive cesium isotopes [1, 2]. However, the reason for the selectivity to sulfate compounds in production mechanism of radioactive aerosols is not clear. Attachment behavior of fission products to solution aerosol particles has been investigated in order to elucidate the production mechanism of radioactive aerosols. The aerosols which contains a radioactive fission product were generated and attachment ratio of fission product aerosol was measured to reveal the production mechanism of the radioactive solution aerosols in the present work.

EXPERIMENTS: The fission-product aerosols were produced by passing a primary solution aerosol through a chamber in which ²⁵²Cf source was placed. The ²⁵²Cf source was covered by two Havar foils which thickness is 2.5 µm each to reduce kinetic energy of fission products emitted from the source. Primary aerosols were generated from alkali halide (NaCl, NaBr, NaI, CsCl) and ammonium sulfate solution of 0.1 wt% concentration using an atomizer. The primary solution aerosol was classified by size using a differential mobility analyzer before the injection to the ²⁵²Cf chamber. The radioactive aerosol was produced in the chamber by attaching fission products to aerosol particles and collected on a polycarbonate filter. Radioactivity of fission products attaching to aerosol particles was measured by gamma-ray spectrometry using a Ge detector. Fission products emitted in the chamber from the ²⁵²Cf source were also collected on a cellulose filter placed directly on the source and measured by the Ge detector.

RESULTS: The attachment ratio of fission products to aerosol particles was estimated from the ratio of photopeak areas for each fission product and shown in Fig. 1 as a function of total surface area of aerosol particles. The attachment ratio seems to be proportionate to the surface area of aerosol particles in the small surface area region. The increasing rate of the relationship varies with materials of the aerosol particles. The proportional relation and the variation of increasing rates suggest that the attachment ratio of fission products to aerosol particles depends on both geometric collision rates and chemical effects. On the other hand, the increasing rates decrease and the attachment ratios were saturated in the larger region of the surface area of aerosol particles. This trend indicates that the attachment behavior of fission products to aerosol particles could be held in an adsorption-desorption equilibrium.

The relationships between the surface area and the attachment ratio for ¹⁰⁴Tc were fitted by a function on the basis of the assumption of the adsorption-desorption equilibrium between fission products and surface of aerosol particles. As results of the fitting which are shown by solid lines in Fig. 1, equilibrium constants and saturated attachment ratios were obtained for the combination of ¹⁰⁴Tc and each solution aerosol. In the comparison among the estimated equilibrium constants of ¹⁰⁴Tc for solution aerosols, it was found that the equilibrium constant for solution aerosols. This difference might be caused by density profiles of the anions in the fine solution particles that is explained by molecular dynamics simulations [3, 4].



Fig. 1 Attachment ratio of ¹⁰⁴Tc to various solution aerosol as a function of total surface area of aerosol particles.

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PR10-2 An Application of Imaging Plates to Size Measurement of Radioactive Aerosol Particles Produced in Accelerator Room Air

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INTRODUCTION: Very fine radiation-induced aerosol particles are often observed in air of accelerator rooms during machine operation. The particles are produced by an air ionization process by primary beams or by charged particles emitted from beam loss points. The particles incorporate radioactive atoms to form radioactive particles. The size for radioactive particles was often measured using a wire screen technique in accelerator facilities. Convenient size measurement techniques are needed from the viewpoint of radiation protection in accelerator facilities.

When aerosol particles pass through a stack of wire screens, a part of the particles are trapped on the screens according to their particle size. As the loss by the screens is expressed as a function of particle size, coarseness of screens, and flow rate of particles, the particle diameter can be calculated by measuring the loss by each screen.

In this work, air irradiation experiment was performed in the FFAG (Fixed Field Alternating Gradient) proton accelerator in the Research Reactor Institute (KURRI), and a combination method of imaging plate (IP) measurement and the wire screen technique was applied to estimate particle size of ¹¹C-bearing aerosols.

EXPERIMENTS: A screen device consisting of a stack of stainless steel wire screens and a PTFE backup filter was used for size measurement of the radioactive aerosol. The stacking order and number of the screens are single 100, 200, 300-mesh screens and 40 pieces of 500-mesh screens in a direction toward the downstream.

An air-irradiation chamber was installed at the upstream of the beam dump of the beam extraction line of the FFAG main ring (Fig. 1). The chamber was irradiated for 1 h with a stable beam of nominal 150-MeV protons. The beam current was 1.8 nA. During the irradiation aerosol-free air was continuously introduced at the flow rate of 8.1 L/min to the chamber, and the irradiated air was sampled with the screen device at the rate of 7.5 L/min. A part of the irradiated air was introduced to an SMPS (Scanning Mobility Particle Sizer) to monitor the number-based aerosol size simultaneously with the screen collection.

In the aerosol the radionuclides affecting the IP exposure are virtually only positron emitters such as ¹⁵O (half life: 2 min), ¹³N (9.965 min) and ¹¹C (20.39 min). After the short-lived ¹⁵O was decayed, a radiation image of selected screens and the backup filter was obtained by

30-min exposure of one single IP (Size 35 x 43 cm). To evaluate contribution of 11 C and 13 N to the intensity of photostimulated luminescence (PSL), a separate air irradiation experiment was performed under the same irradiation and sampling conditions. After the irradiation the decay of activity for the screen and the backup filter was analyzed using GM detectors.

In comparison with ¹¹C, the 478-keV photopeak for ⁷Be was analyzed using Ge detectors to obtain size distribution for ⁷Be-bearing aerosol particles.



Fig. 1 Irradiation Chamber installed in the beam extraction line

RESULTS: The ¹¹C activities for each screen and the backup filter were estimated from the decay analysis mentioned above. The particle size of ¹³N-bearing particles was not analyzed in this work because ¹³N gaseous species like nitric acid interferes with collection of the ¹³N-bearing particles [1]. The activity ratio of particles penetrating the screens to whole particles before penetrating the screens is governed by their diffusion coefficient expressed by a function of particle size. The size distribution was calculated by fitting the curve of the penetrating ratios vs. the number of screens to the theoretical equation [2]. Log-normal distributions are assumed as the size distribution function in the calculation because the number-based size distribution was confirmed to be log-normal by the SMPS measurement.

The geometric mean diameter, d_g , for the ¹¹C-bearing aerosol particles was successfully obtained using the combination way of the wire screen collection and the IP measurement. The diameter was found to be 68.0 nm for the ¹¹C-bearing particles, while that for ⁷Be-bearing particles obtained by the Ge detector measurement was 63.4 nm. The activity-based geometric mean diameters were larger than the number-based one by ca. 20 nm. This shows that attachment process of radioactive atoms to the surface of aerosol particles is included in the radioactive particle formation.

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PR13 Project Research on a Study on Biological Character and use of the Particle Induced by the Boron Neutron Capture Reaction

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Objectives and Participating Research Subjects

In this project, we are intending to develop the new application using the characteristics of the particles from the neutron capture reaction.

PRS-1 Analysis of mutation in the mammalian cells induced by BNCR (boron neutron capture reaction)

(Y. Kinashi et al.)

- PRS-2 Analysis of double strand breaks in the mammalian cells induced by BNCR (S.Takahashi *et al.*)
- PRS-3 Development of the PARP repressor reinforced in its function by BNCR (Y.Uto *et al.*)
- PRS-4 Development of the model animal shouing the blood vessel damage by BNCR(R. Wate *et al.*)

Main Results and Contents

PRS-1 studied gamma-H2AX focus data and investigated the DNA-dsb (double strand breaks) damage and association of the mutation induction following the neutron radiation beam used for BNCT in KUR. The mutagenic effects of BNCT on the normal tissue cells that do not take up the boron compounds may cause the genetic instability and second cancer decade years after BNCT. The mutagenicity measured by the frequency of mutations induced by neutron irradiation with or without boron compound. The HPRT locus was

examined in Chinese hamster ovary (CHO) cells irradiated with neutrons of KUR.

The frequency of mutations after neutron irradiation with 10ppm BPA was about 5 times and more increased than 1Gy gamma-ray irradiation. the gamma-H2AX foci size at after 1Gy of neutron irradiation with 10ppm BPA. After 2 hours of neutron irradiation, about 20% of cells had big foci,

The result suggested that The mutation induction might be increased due to the inferiority of the ability of DNA damage repair following BNCR.

PRS-2 was not able to carry out its experiments because KUR (Kyoto University Research Reactor)

has been stopping during this year.

PRS-3 was not able to carry out its experiments because KUR has been stopping during this year.

PRS-4 was not able to carry out its experiments because KUR has been stopping during this year.

PR13-1 Association of the DNA-double Strand Breaks Damage and the Mutation Induction Following the Neutron Radiation Beam used for BNCT

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INTRODUCTION: We have previously reported the increased mutagenicity of thermal neutrons and the dose rate effect of the neutron radiation beam [1]. Clinically, the mutagenic effects of BNCT on the normal tissue cells that do not take up the boron compounds may cause the genetic instability and second cancer decade years after BNCT. In this study, we analyzed gamma-H2AXfocus data and investigated the DNA-dsb (double strand breaks) damage and association of the mutation induction following the neutron radiation beam used for BNCT in Kyoto University Research Reactor (KUR).

MATERIALS & METHODS: The mutagenicity measured by the frequency of mutations induced by neutron irradiation with or without boron compound. The hypoxanthine-guanin-phosphoribosyl-transferase (HPRT) locus was examined in Chinese hamster ovary (CHO) cells irradiated with neutrons of KUR. A stock solution of 10B-para-boronophenylanine (BPA) was used for this experiment. Cell suspensions were incubated with BPA at 10 ppm concentration 1 hour before neutron irradiation. To determine mutation frequencies, each treated culture was incubated with non-selective medium for 7-9 days to allow phenotype expression. Then, $2x10^5$ cells were added to each dish containing 6-thioguanine and incubated for 10-14 days, after which time the mutant colonies were counted. The mutation frequency is expressed as the number of resistant colonies divided by the total number of viable cells as determined by cloning efficiency at the time of selection. DNA double-strand breaks (DSBs) in CHO cells following neutron irradiation were detected using the gamma H2AX foci assay. Immunofluorescence staining was performed as described previously (2). Briefly, irradiated cells were incubated 30 min.or 120 min., and washed with cold PBS. After fixation with 3.6% formalin Immunofluorescence images were acquired using a fluorescence microscope (KEYENCE, BZ-9000), and then analyzed using the BZ-9000 optional software and Image-J (National Institutes of Health).

RESULTS and DISCUSSION: Table 1 shows the mutation frequency in the HPRT locus in CHO cells after 1Gy neutron irradiation. The frequency of mutations after neutron irradiation with 10ppm BPA was about 5 times and more increased than 1Gy gamma-ray irradiation.

Table 1 HPRT mutation frequencies of CHO cells ex
posed to 1Gy of gamma-rays, neutron and BNCR

	Gamma ray	Neutron	BNCR (10ppm BPA)
HPRT mutation frequency / 105 cells	1.2 ± 1.5	3.2±2.5	6.8±3.5

Figure 1 shows the gamma-H2AX foci size at after 1Gy of neutron irradiation with 10ppm BPA. After 2 hours of neutron irradiation, about 20% of cells had big foci (>5.0 μ m2), On the other hand, the cell with the big focus was less than 1% at 2 hours after gamma ray irradiation.



0025 025:05 05:10 10:15 15:2 2:25 25:3 30:40 40:50 >50 Fig.1 gamma-H2AX foci size at 30min. (white bars) (μm^2) and 120min. (black bars) after 1Gy of neutron irradiation with 10ppm BPA.

In this study, we found that the large foci which are a serious DNA damage increased by BNCR. The mutation induction might be increased due to the inferiority of the ability of DNA damage repair following BNCR.

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I-2. Collaboration Researches

- 1. Slow Neutron Physics and Neutron Scattering
- 2. Nuclear Physics and Nuclear Data
- 3. Reactor Physics and Reactor Engineering
- 4. Material Science and Radiation Effects
- 5. Geochemistry and Environmental Science
- 6. Life Science and Medical Science
- 7. Neutron Capture Therapy
- 8. Neutron Radiography and Radiation Application
- 9. TRU and Nuclear Chemistry
- 12. Others

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INTRODUCTION: The B-3 beam port of Kyoto University Research Reactor (KUR) had long been used as a four-circle single-crystal neutron diffractometer (4CND). For the last decade, however, the 4CND was so old that its research activity on neutron science was quite low. Therefore, the 4CND needed to be replaced and a new neutron diffractometer has been required at the B-3 beam port. Also, the new neutron diffractometer (Compact multipurpose neutron diffractometer) is critical for the structural investigations of energy storage materials such as hydrogen absorbing alloys and rechargeable lithium-ion batteries. The neutron (powder) diffraction is a powerful tool to determine the positions of light elements (e.g., hydrogen and lithium) in solids. Here, we report the current status of the B-3 beam port of KUR and the preliminary neutron diffraction experiments using a hydrogen absorbing alloy.

INSTRUMENT: The compact multipurpose neutron diffractometer is now being installed on the B–3 beam port. The neutron wavelength (λ), which is monochromatized by the (220) plane of a Cu single crystal, is 1 Å. To cover the detector area (6 ° $\leq 2\theta \leq 150$ °), 24 ³He tube detectors (1/2 inch in diameter) have been prepared. The distances from the monochromator to the sample and from the sample to the detector will be 1.9 m and 1.2 m, respectively. A detector bank including the 24 ³He tube detectors will be placed on an arm of the goniometer. In addition, the new beam shutter was installed at the B-3 beam port.

PRELIMINARY EXPERIMENTS: The preliminary neutron diffraction experiments were performed using the hydrogen absorbing alloy, $(Ti_{0.31}Cr_{0.33}V_{0.36})D_{1.7}$, where D is the deuterium. It is worth noting that $Ti_{0.31}Cr_{0.33}V_{0.36}$ is a null alloy for neutron scattering due to its atomic compositions (i.e., $b_c[Ti_{0.31}Cr_{0.33}V_{0.36}] = 0$; $b_c[Ti] = -3.370$ fm, $b_c[Cr] = 3.635$ fm, and $b_c[V] = -0.443$ fm), where b_c is the coherent scattering length. The crystal structure has been refined on the basis of a cubic CaF₂-type structure with a = 4.288(1) Å, using X-ray diffraction with Cu K α radiation (see Fig. 1(b)). As shown in Fig. 1(a), we succeeded to observe several Bragg reflections for the $(Ti_{0.31}Cr_{0.33}V_{0.36})D_{1.7}$ on the B–3 beam port; which could be indexed on the basis of $\lambda = 1$ Å. Note that the Bragg reflections correspond to the D–D correlations because of the null alloy.

Furthermore, the data acquisition group of the neutron science division of KEK (KEK–KENS DAQ group) has used the B–3 beam port to assess their new ⁶Li-glass neutron detector system, LiTA12. The LiTA12 system consists of a ⁶Li-glass neutron detector with a multianode photo multiplier tube (MA–PMT), an amplifier, and an analog-to-digital converter (ADC) board. The B–3 beam port has a wide space around the sample position; therefore we can easily install any other system like the LiTA12 system.

IMPROVEMENT OF MONOCHROMATOR STAGE: The old Cu monochromator stage of the 4CND was removed (Fig. 2(a)), and then the new Cu monochromator stage was installed at the B–3 beam port (Fig. 2(b)). The Cu monochromator stage consists of two goniometers: rotation and swivel stages (RA07A-W and SA05B-RM, Kohzu Precision Co., Ltd.).





(a)

(b)



Fig. 2. Improvement of Cu monochromator stage: (a) old stage, and (b) new stage, respectively.

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INTRODUCTION: Spiders trap insects using a web net. The fact that they eat them without chewing, indicate that spiders possesses highly efficient digestive enzymes [1]. Our previous studies suggest that spider's proteolytic enzymes are able to digest synthetic spider dragline amyloid fibers [2,3]. Thus, the spider protease has the potential ability to digest amyloid fibers including pathogenic β -amyloid, such as amyloid fibrils, that are associated with the development of Alzheimer's disease [1]. Therefore, we purified, identified, and characterized the enzymes derived from *Nephila Clavata*. The sequence analyses of the blotted protein on a PVDF membrane showed N-terminal amino acid residues. Based on this sequence information, we tried to clone the protease gene.

EXPERIMENTS: The spiders (*Nephila Clavata*) were collected in the field of Kyoto University Research Reactor Institute and the spider saliva including proteolytic enzymes was obtained from *Nephila Clavata* (50 heads) by electrical stimulation. The extracts were stored in a deep freezer until use.

Casein protease assay [4] of the crude spider enzymes was performed, as previously reported [1]. Peptide substrates for the assay of the protease activity of spider enzymes were chemically synthesized by the Boc solid phase method, treated with hydrogen fluoride, and purified by reversed phase HPLC [1, 4].

Based on the N-terminal sequence, evidenced by the Edoman's degradation method, we prepared generated primers for the gene cloning and RT-PCR was performed using mRNA's from gut cells or full bodies of *Nephila Clavata*.

RESULTS: Spider's digestive fluid that includes proteolytic enzymes was obtained from *Nephila Clavata* by electrical stimulation using a micropipet. The extracts were applied to SDS-PAGE and the enzymatic activity of the protein bands was estimated by a casein protease assay [4]. Two protein bands showed protease activities and their molecular weights were estimated to be approximately 21.9 and 19.5 kDa, based on the SDS-PAGE analysis, as we determined previously [1].

To determine the gene sequence of the spider protease,

we tried to obtain its mRNA. For this purpose, generated primers for the gene cloning of the protease were specifically designed and RT-PCR was performed using mRNA from gut cells or full bodies of *Nephila Clavata*. Several clones were isolated and provided second PCR to produce cDNA's of spider proteases. The purified candidate cDNA's were applied to PCR again, then their cDNA sequence were directly determined. However, the sequence analysis of the cloned spider protease gene revealed that the cloned genes were non-specifically amplified during RT-PCR. Therefore, generating primers were re-designed by considering codon bias of spider. RT-PCR and following PCR produced several candidate cDNA's . Their sequence analyses is now in progress.

We also prepared the cDNA of cocoonase to estimate amyloid digestive activity and expressed it using *E. coli* cells. Recombinant cocoonase was successfully over-expressed as a inclusion body. The protein was purified and analyzed by reversed phase HPLC and MALD-TOF/MS, respectively. To activate the protein, refolding reactions were carried out under several conditions and its protease activity will be estimated using a synthetic substrates.

In conclusion, Spider protease was extracted from its digestive fluid and exhibited strong protease activity. The protease can be classified as a Ca^{2+} -dependent carboxypeptidase, based on the results of protease inhibition assays. Spider genes were amplified using generating primers, base on the N-terminal amino acid sequence, by RT-PCR and determined by the direct sequencing method. However, DNA sequence analysis indicated that the cloned DNA was non-specifically amplified during RT-PCR. The cloning of spider protease genes is in progress. In addition, we also prepared the cDNA of the protease, cocoonase andover-expressed it using E. coli expression system. Recombinant cocoonase was successfully refolded as a soluble protein. Its biological activity will be estimated and provide a critical insight to understand the amyloid digestive activity of spider and Bombyx mori.

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CO1-3 Development of a Plano-elliptical Supermirror with Metallic Substrates

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INTRODUCTION: Progress of neutron optical devices is very significant, however, it is still very difficult for neutron aspherical focusing mirror. The metallic substrate is robust and ductile, able to withstand high radiation irradiation and high-temperature. It is therefore possible to install them even at a place close to the neutron target and moderator. Furthermore, it is possible to fabricate a large focusing supermirror by multiple seg-mented combining mirrors with mechanical fastening entailing the usage of screw holes and fixture tabs. The big problem is required surface roughness for neutron mirror. It is smaller than 0.5 nm even for m=3 supermirror coating and the mirror size is large. Here m is the maximum crit-ical angle of the mirror in units of critical angle of natural nickel. In order to realize large ellipsoid neutron focusing supermirror, we are doing a lot of trials. In this study, we show the neutron reflectivity of a large planoelliptical supermirror with metallic substrates.

EXPERIMENTS: We fabricated a large plano-elliptical metallic substrates with electroless nickel-phosphorus (NiP) plating, based on the technology using ultrahigh precision cutting with correction processing, followed by mechanical precision polishing. We manufactured a two segment large plano-elliptical supermirror with m=3 supermirror coating for the Soft Interface Analyzer (SO-FIA) at the Materials and Life Science Experimental Facility (MLF) of Japan Particle Accelerator Research Complex J-PARC [1]. Because the area limitations of machines used for manufacturing and supermirror coat-ing. To realize sample focusing optics, the focusing mir-ror was installed at an equidistant position between first slit and the sample position. One focal point was at the first slit. The other was at the sample position. The major axis of the mirror was designed to be 2150 mm. The mi-nor axis was designed to reflect neutrons to use the typi-cal wavelength band (> 0.2 nm) for reflectivity measure-ments at SOFIA. For the mirror coating, the m=3 super-mirror was chosen to keep as high a reflection angle as possible while the neutron simultaneously keeping reflec-tivity sufficiently high. Therefore, the minor axis was chosen as 21.5 mm to reflect the neutrons on the mirror with an incident angle of 10 mrad. The mirror length was chosen as 550 mm to avoid interference with other de-vices installed to SOFIA. The width was 60 mm, as de-termined from the maximum beam size width of 50 mm. The manufacturing, polishing and cleaning of the metallic substrate were conducted at RIKEN [2]. The supermirror coating was conducted with KUR-IBS [3]. The neutron experiments were conducted at the SOFIA reflectometer at J-PARC/MLF.

RESULTS: Figure1 shows neutron reflectivity of the 10 measured locations in the plano-elliptical supermirror with metallic substrates. Typical surface roughness of the top surfaces of metallic substrates was approximately 0.3 nm rms using a white light interferometer (NewView 7200; Zygo Corp.).Using the KUR-IBS, m=3 NiC/Ti supermirrors were coated onto the substrates surface. The maximum neutron reflectivity of the supermirror on the elliptical metallic substrates was 0.8, which is almost identical to that with flat glass substrates. However, the average of neutron reflectivity was not so high because of surface contamination resulting from inadequate cleaning.

The figure error shape was $4.6 \,\mu\text{m}$ P–V. The slope error was 33 μ rad rms. The focusing supermirror was installed at the SOFIA neutron reflectometer, showing high neutron reflectivity and giving minimal beam width of 0.34 mm in FWHM. Because of the large beam divergence accepted by the mirror, the count rate with the focusing mirror was 3.3 times higher than that obtained using conventional two-slit collimation[2].



Fig. 1. Neutron reflectivity of the 10 measured locations in the plano-elliptical supermirror with metallic substrates. The focusing mirror consists of two segments (Mirror A and B).

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CO1-4 Neutron Irradiation Effect of High-density MoO₃ Pellets for Mo-99 Production (3)

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INTRODUCTION: JAEA has a plan to produce ⁹⁹Mo by (n, γ) method $((n, \gamma)^{99}$ Mo production), a parent nuclide of ^{99m}Tc. In 2014, irradiation test was carried out with the high-density molybdenum trioxide (MoO₃) pellets in the hydraulic conveyer (HYD) of the Kyoto University Research Reactor (KUR) and the ^{99m}Tc solution was extracted from the solution of irradiated MoO₃) pellets. In this study, the qualification of ^{99m}Tc solution was evaluated.

EXPERIMENTS: The high-density MoO₃ pellets were irradiated in the HYD of the KUR. After the irradiation tests, the irradiated MoO₃ pellets were transported from KUR to JMTR-HL. The irradiated pellets were dissolved with 6M-NaOH solution (Mo Solution) in the Lead Cell and the ^{99m}Tc was extracted from the Mo solution by the solvent extraction method using methyl ethyl ketone (MEK). Two types of the alumina columns were prepared for the extraction of ^{99m}Tc solution. At first, MEK containing ^{99m}Tc was passed through a basic alumina column for removing impurities such as Mo and H₂O. Next, the MEK was flown through the acidic alumina column in order to lead the whole ^{99m}Tc. Finally, saline (0.9%-NaCl) was passed the acidic alumina column and ^{99m}Tc solution was collected as a product. The inspection of ^{99m}Tc solution was carried out using pH meter, ICP-AES, HPLC, germanium detector, and so on.

RESULTS: After the dissolution of irradiated MoO₃ pellets and extraction of ^{99m}Tc solution, ⁹⁹Mo and ^{99m}Tc activities were measured in the Mo solution and ^{99m}Tc solution by the germanium detector. Table 1 shows the result of ^{99m}Tc recovery rates from the Mo solution ^[1]. The recovery rate of the 1st and 2nd runs achieved the target values (>80%).

 Table 1
 99m Tc recovery rates from the Mo solution

No.	Activity of 99mTc (Bq)	Recovery rate (%)
1 st run	1.09×10^{9}	82.3
2 nd run	$8.53 imes 10^8$	95.7

Table 2 shows the qualification of ^{99m}Tc solutions. In this table, the standard values were decided under the framework of Strategic Program for Basic Nuclear Research launched by the MEXT. Radionuclidic purities and radiochemical impurities were measured and the values obtained satisfactory results. Especially, ^{92m}Nb observed in the Mo solution as impurities, but ^{92m}Nb was

not detected in the ^{99m}Tc solutions of 1st run and 2nd run. Figure 1 shows the γ -ray spectrum of Mo solution and ^{99m}Tc solution. MEK content was lower than that of the standard values (5,000ppm). Osmotic pressure of the ^{99m}Tc solutions was almost the same values of that of the saline and endotoxin was not detected. From the results, the high purity ^{99m}Tc solutions were obtained by this method.

Table 2 Qualification of ^{99m}Tc solutions

Tuble 2 Quantication of Te Solutions				
Sample	Standard Values	1 st run	2 nd run	
pН	4.5 - 7.0	5.5	5 - 6	
Radionuclidic purities	≦0.015%	99mTc(only)	99mTc(only)	
Radiochemical impurities	≦5(%)	2.0	-	
MEK content (ppm)	\leq 5000	404	-	
Al content (ppm)	≤ 10	-	<5	
Osmotic pressure (mosm)	270 - 300	286	297	
Endotoxin	N.D.	N.D.	-	



Figure 1 γ -ray spectrum of Mo and ^{99m}Tc solutions.

CONCLUSION: After the irradiation test of the high-density MoO₃ pellets in the KUR, 99m Tc was extracted from the Mo solution. The qualification of 99m Tc solution by the solvent extraction method was satisfied the standard values for 99m Tc radiopharmaceutical products.

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CO1-5 Development of High Spatial Resolution Cold / Ultra-cold Neutron Detector

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INTRODUCTION: We have been developing cold/ultra-cold neutron detectors with spatial resolution of several ten nanometers, by using fine-grained nuclear emulsion [1, 2] with its AgBrI crystal size of 40nm, and nuclides which emit heavily ionizing particles after absorbing neutrons with high absorption cross section. One of the aim is to measure the position distribution of ultra-cold neutrons in the quantized states in the earth's gravitational field for the test of gravitational inverse-square law and equality of gravitational and inertial mass, as done in previous studies [3, 4], but more precisely, by 1 or 2 orders of magnitude.

The extremely high resolution of several ten nanometers should be realized with a 10nm-thick ¹⁰B layer, coated with nuclear emulsion. When a neutron is absorbed in the ¹⁰B layer, an alpha particle and a ⁷Li will be emitted. After the development, emulsion layer will be observed with an optical microscope. A track of an alpha particle or a ⁷Li starting from the absorber layer will be seen in the emulsion layer just next to the absorber layer. To decide the absorption point, the track will be extrapolated into the absorber layer. Then, ambiguity of the absorption position will be limited by the thickness of the absorber.

In this experiment, we made the detector and exposed to cold neutrons to test the detection principle.

EXPERIMENTS: A detector was made in the following procedure. A layer of ¹⁰B with thickness of 10nm was sputtered on a base, a piece of silicon wafer, at KURRI by M. Hino. The ¹⁰B layer was coated with fine-grained nuclear emulsion melted at the temperature of 40 degrees centigrade. Thickness of the emulsion layer coated was 10micron. After drying, it was packed in a light-tight bag made of thin layers of aluminum and polyethylene.

The detector was exposed to cold neutrons with energy of 10meV in average at BL05 in J-PARC MLF. After the development, observation was done by an optical microscope with an epi-illumination system.

RESULTS: As shown in Fig. 1, tracks from absorption events were clearly seen at the depth around the surface of the 10 B layer.



Fig.1. Tracks from absorption of neutrons by ¹⁰B are surrounded by red ellipses.

But, we found tracks from absorptions by ¹⁰B also inside the emulsion layer apart from the ¹⁰B layer. This showed that some of ¹⁰B moved into emulsion layer. ¹⁰B layer turned out to be instable when it is coated directly by nuclear emulsion.

We are now developing methods to stabilize it.

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CO2-1 Research and Development for Accracy Improvement of Neutron Nuclear Data on Long-lived Radioactive Nuclei at KURRI-Linac

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INTRODUCTION: There are strong requests for reducing the uncertainty of neutron capture cross section data of minor actinides (MAs) to estimate the transmutation rate of those long-lived radioactive nuclei in the innovative reactor system. In recent years, intense pulsed spallation neutron sources such as J-PARC[1], n-TOF[2] and LANSCE[3] facilities became available to remarkably improve the precision of neutron TOF data. However, there are discrepancies out of a range of tolerance between current experimental results. It is understood that the unrecognized systematic errors make a difference. In order to recognize and reduce the systematic errors, the project entitled as "Research and development for Accuracy Improvement of neutron nuclear data on Minor ACtinides (AIMAC)" has been started. In this project, we aim at obtaining the resonance parameters precisely of MAs by combining the neutron capture γ -ray measurement to transmission neutron measurement at KURRI-Linac. In this year, new detection system of neutron capture γ rays and transmission neutron was constructed and its performance was confirmed.

EXPERIMENTS: We constructed 4π bismuth germinate (BGO) scintillation detectors composed of 12 BGO cylindrical crystals having 2 inch. in diameter and 2 inch. in length for measurement of neutron capture γ rays. On the other hands, a 6 mm thick GS20 ⁶Li-glass scintillator was used as a transmission neutron detector.

In the performance test, we used a sealed MA sample of ²³⁷Np. Neptunium oxide powder of 1.13 g packed in an aluminum disk container of 30 mm in diameter and 0.4 mm thick wall. The sample was placed in the geometrical center of the BGO detectors using an aluminum sample folder. To decrease the constant background due to decay γ rays from radioactivity (26 MBq) of ²³⁷Np, a 3 mm thick lead shield was inserted in the sample folder and BGO detectors. A distance between the sample and the KURRI-LINAC neutron source was 10 m. To obtain the incident neutron spectrum, a ¹⁰B sample was also used. Transmission neutron spectra with and without the ²³⁷Np sample were measured by the ⁶Li-glass detector.

RESULTS: Preliminary result of neutron capture cross section of ²³⁷Np is shown in Fig. 1. The relative cross sections were normalized to the evaluated value of JENDL-4.0[4] at 0.0253 eV. A correction of neutron multiple-scattering and self-shielding in the sample has not be made yet. Although the amount of measuring time is about one hour, enough net counts were obtained in the resonance region. Comparison of transmission neutron

spectra with and without ²³⁷Np sample by the ⁶Li-glass detector is shown in Fig. 2. We could observe the significant differences between those spectra with a high signal to noise ratio..

The results show that the developed capture γ -ray and transmission neutron detection system has a high performance to determine the resonance parameters.



Fig.1 Preliminary results of neutron capture cross section of ^{237}Np



Fig.2 Comparison of transmission neutron spectra with and without ²³⁷Np sample by the ⁶Li-glass detector

Present study includes the result of "Research and Development for accuracy improvement of neutron nuclear data on minor actinides" entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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CO2-2 Activation Measurements of Neptunium-237 at KURRI-Linac

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INTRODUCTION: A series of experiments with an activation method has been performed to measure thermal-neutron capture cross-sections of Minor Actinides (MAs) under the project entitled by "Research and development for <u>Accuracy Improvement of neutron nuclear</u> data on <u>Minor AC</u>tinides (AIMAC)". Neptunium-237 is one of important MAs because it contributes to the long-term radiotoxicity of nuclear wastes. Then, the present work performed cross-section measurements of ²³⁷Np by an activation method with a neutron source generated by the KURRI-Linac.

EXPERIMENTS: A standardized solution of 237 Np (4 kBq) was pipetted on a quartz plate (10mm × 15mm × 2mm^t), and dried by an infrared lamp. Then, the Np sample was put into an aluminum target holder together with Co and Au foils as neutron monitors. The target holder was set near the moderator tank at the target room of KURRI-Linac. The irradiation was performed for 14 hours. The KURRI-Linac was operated under the condition: repetition rate 50Hz, pulse width 4µs, 108µA beam current and 3-kW power.

After the irradiation, induced activities of the samples were measured with a high purity Ge detector. **Figure 1** shows an example of γ -ray spectrum of the irradiated ²³⁷Np sample.



Fig.1 γ -ray spectrum of the irradiated ²³⁷Np sample

RESULTS and DISCUSSION: Neutron flux components were derived from the induced activities of the monitors on the basis of the Westcott's convention[1]. The thermal-neutron flux was $2.97 \times 10^8 (n/cm^2s)$ at the irradiation position of 3-kW operation. The epi-thermal

index was 0.053. The reaction rate of the ²³⁷Np sample was calculated with 312-keV and 984-keV γ -ray yields, detection efficiencies, decay data and γ -ray emission probabilities. The reaction rate was $(7.12\pm0.25)\times10^{-14}$ (1/s). The experimental reaction rate was compared with the calculated one with evaluated nuclear data of ²³⁷Np. The neutron flux distribution at the irradiation position was calculated by MVP2.0[2] with JENDL-4.0[3], and plotted in **Fig.2**. The calculated flux was normalized with the reaction rate of Au monitor. With the normalized neutron flux and the cross-section data ²³⁷Np from JENDL-4.0[3], the calculated reaction rate resulted in (7.41±0.14)×10⁻¹⁴ (1/s). The calculated reaction rate was good agreement with the experimental one within the limits of error.



Fig.2 Neutron flux distribution calculated by MVP Codes

CONCLUSION: Since the thermal-neutron flux was found to be 10^8 (n/cm²s) order, activation measurements would be possible under adequate experimental conditions: irradiation time, repetition rate, beam current and so on. The activation measurement of ²³⁷Np was performed. It was found that this measurement supported the evaluated data of JENDL-4.0.

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Development of the Epi-Thermal Neutron Measurement Method Using a Thick Boron Sample

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INTRODUCTION: Evaluation of neutron fluence and neutron dose equivalent for the epi-thermal neutron region is very important in work places with neutron sources or nuclear fuels as well as irradiation fields in a boron neutron capture therapy. The epi-thermal neutrons are usually measured using a gold activation detector, a gas proportional counter or a scintillator. However, it is not easy to determine precisely the neutron fluence for the epi-thermal neutrons in an irradiation field because of large uncertainty of reaction cross sections in the epi-thermal region. In the present study, we have developed an epi-thermal measurement method that is not affected by nuclear reaction cross sections. We also developed an epi-thermal neutron camera consisting of GEMs and resonance filters for neutrons up to 10 keV.

EXPERIMENTS: A collimated neutron beam was obtained by the photo neutron reaction using a water-cooled tantalum target at the KURRI Linac [1]. Characteristics of a neutron detection system composed of a 6Li6natGd10B3O9:Ce+ (LGB) scintillator and an NaI(Tl) scintillator were experimentally evaluated by means of the time-of-flight (TOF) method. The 50 mm-diameter and 5-mm thick LGB scintillator was set at the center of the beam line. The 76.2 mm-diameter and 76.2 mm thick NaI(Tl) was located out of neutron beam at an angle of 135 or 90 degrees with respect to the neutron beam direction. When the LGB scintillator detects neutrons by the ${}^{10}B(n,\gamma\alpha)$ reaction, 478 keV monoenergetic gamma rays are produced and subsequently detected with the NaI(Tl) scintillator. Moreover, the absolute neutron fluence is determined by measuring gamma rays from the ${}^{10}B(n,\gamma\alpha)$ reaction with the NaI(Tl) scintillator in setting a 5-cm thick ^{nat}B total absorption sample in front of the LGB scintillator. Number of alpha particles and gamma rays produced by the ${}^{10}B(n,\gamma\alpha)$ reaction in the LGB scintillator are absolutely determined by the coincidence measurements. Uncertainty caused by the

cross sections of the ${}^{10}B(n,\alpha\gamma)$ reaction is reduced by using the boron total absorption sample.

RESULTS: Fig.1(a) and (b) show ratio of the pulse height spectra and the TOF spectra of the NaI(Tl) detector with and without the thick ^{nat}B total absorption sample. In Fig.1(a), 478 keV gamma rays due to the ¹⁰B(n, $\alpha\gamma$) reaction is detected around 600 channel. These figures indicate the relative ratio of count rate of the NaI(Tl) scintillator with and without the boron total absorption sample. In the measurements, it was confirmed that the present detection system derived the neutron fluence in the thermal and epi-thermal region.The experimental results will be basic data in order to develop a novel portable epi-thermal neutron detector.



Fig.1. Ratio of (a) the pulse height spectra and (b) the TOF spectra of the NaI(Tl) detector with and without the thick ^{nat}B total absorption sample.

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CO2-4 Measurements of Americium Isotopes by Activation Method at KURRI-Linac

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INTRODUCTION: Neutron capture cross section measurements have been conducted for Minor Actinides (MAs) under the research project entitled by "Research and development for <u>Accuracy Improvement of neutron nuclear data on Minor ACtinides (AIMAC)"</u>.

Since americium-241 and -243 have relatively long half-life as 432yr and 7370yr respectively, these isotopes are important nuclides among MAs in terms of environment load reduction. This is why these two americium isotopes were selected in the present measurements.

EXPERIMENTS: Americium-241, 243 samples were sealed on Al plates with 22mm in diameter and 0.5mm in thickness, respectively. Their active areas were 20mm in diameter. The amounts of the samples were 583kBq for ²⁴¹Am, and 2MBq for ²⁴³Am, respectively. Each Am sample was packed into an Al target holder. Then, sets of monitors of Au and Co foils were sealed at adequate positions on the Al holders. Then, the target holders were set near the moderator tank at the target room of KUR-RI-Linac.

Figure 1 shows the irradiation set-up of the ²⁴³Am target near the moderator tank at the target room of KUR-RI-Linac. The ²⁴¹Am target was irradiated for 35 hours under beam conditions: repetition rate 100pps, beam width 4 μ s, beam current 220 μ A, and 6-kW power operation, while the ²⁴³Am target was irradiated for 40 hours under another beam conditions: repetition rate 50pps, beam width 2.5 μ s, current 67 μ A, and 2-kW power operation.



Fig. 1 Set-up of the ²⁴³Am target at the target room of KURRI-Linac

After the irradiations, induced activities of the samples were measured with an α -ray spectrometer, and a high purity Ge detector. **Figure 2** shows an example of α -ray spectrum obtained by measurements of the irradiated ²⁴¹Am sample for 11 days. On the other hand, the ²⁴³Am sample was measured by the Ge detector. The γ -ray spec-

trum of the 243 Am sample was measured for 20 hours, and is shown in **Fig.3**.







Fig.3 An example of γ -ray spectrum of the irradiated ²⁴³Am sample

ANALYSIS and RESULTS: As for the ²⁴¹Am experiment, the neutron flux was approximately 1.4×10^8 (n/cm²s) on the basis of the Westcott's convention. From the peak counts of ²⁴¹Am and ²⁴²Cm in Fig.2, the rate of ²⁴¹Am(n, γ)^{242g}Am reaction was obtained as (8.63±0.80) ×10⁻¹⁴(/s). As for the ²⁴³Am experiment, the neutron flux was approximately 9.0×10^7 (n/cm²s) at the target position. The decay γ -rays of 744 and 898 keV of ^{244g}Am (10.1h) were observed as shown in Fig.3. The target amount of the ²⁴³Am sample was obtained with the yields of 228 keV γ -ray emitted from ²³⁹Np in equilibrium with ²⁴³Am. From the ratio of γ -ray yields of 744 keV of ^{244g}Am and 228 keV of ²³⁹Np, the ²⁴³Am(n, γ)^{244g}Am reaction rate was obtained as (1.14±0.03)×10⁻¹⁵(/s), which only included statistical errors.

Furthermore, detailed analyses are underway for the ²⁴¹Am and ²⁴³Am experiments carried out at KUR-RI-Linac.

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Present study includes the result of "Research and Development for accuracy improvement of neutron nuclear data on minor actinides" entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

CO3-1 Development of In-reactor Observation System Using Cherenkov Light (VII)

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INTRODUCTION: On-line surveillance system which can visualize and quantitatively evaluate reactor statuses will contribute to reactor operation management. Development of an on-line reactor core imaging system using Cherenkov light was started in 2009. Previously, the total reactor power of the KUR was successfully estimated from RGB brightness components of the Cherenkov light at the core using a commercial CCD camera^[1]. On the other hand, significant difference of changing rate of brightness with total reactor thermal power was observed between the TC and D2O side of a fuel element^[2]. In this study, in order to assess of applicability of Cherenkov light to acquisition of the reactor thermal power distribution, the image brightness of the Cherenkov light from several fuel elements was compared to the thermal power calculated by SRAC code system^[3].

EXPERIMENTS: The CCD camera (AEC-100ZL, Q·I Inc.) has been inserted into core-observation pipe of KUR during increasing the reactor power thermal from 1 to 5 MW. The output of the CCD camera has been collected as a movie file by a video recorder. Figure 1 shows the CCD camera image of the core of KUR. The Cherenkov light from the fuel elements of the 3rd to 7th column at the "Ho" row^[4] has been analyzed. Since the G and B brightness components have caused halation at the higher reactor thermal power than about 1 MW, the R component has been used to correct the total image brightness.

RESULTS: Table 1 shows the comparison of the thermal power of the fuel elements estimated from the



Fig. 1. The CCD camera image of the core of KUR. The fuel elements of the 3^{rd} to 7^{th} column at the "Ho" row were indicated by a red box.

brightness of the Cherenkov light with the thermal power calculated from the SRAC code system. The comparison was performed at 1 and 5 MW of the total reactor thermal power. From the results, for the "Ho"-3, 4, and 5 fuel elements, the thermal power evaluated from Cherenkov light was almost the same as the one calculated from SRAC code. On the other hand, for the "Ho"-6 and 7 fuel elements, about 15 % and 35% differences were observed, respectively. The possible causes were that shadow of the equipment around the core and/or anisotropic emission of Cherenkov light affected on the brightness of the CCD images. In order to improve the estimation accuracy, installation of a number of cameras at various positions and analysis of the image brightness with taking account of emission angle of Cherenkov light.

CONCLUSION: As part of the development of the visible on-line core surveillance system, the thermal power estimated by brightness of Cherenkov light was compared with the one calculated by the SRAC code system. The results indicate that installation of a number of cameras at various positions and analysis of the image brightness with taking account of emission angle of Cherenkov light are required to precisely obtain the distribution of the thermal power by analysis of Cherenkov light.

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	Reactor thermal	Thermal power of fuel element [kW]		Cherenkov light
	power [MW]	Cherenkov light	SRAC	SRAC
"IIo" 2	1.0	34.3	35.7	0.96
"Ho"-3	5.0	175.2	178.0	0.98
"Но"-4	1.0	41.2	40.8	1.01
	5.0	200.0	203.3	0.98
"IIo" 5	1.0	44.8	41.3	1.09
по -3	5.0	196.7	205.4	0.96
"Но"-6	1.0	31.9	38.4	0.83
	5.0	164.9	190.8	0.86
(III. 22 7	1.0	21.2	31.1	0.68
по -/	5.0	100.3	154.6	0.65

Table 1. Comparison of the thermal power of the fuel elements estimated from the brightness of the Cherenkov light with the thermal power calculated from the SRAC code system.

CO4-1

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INTRODUCTION: Ionic liquid (IL) is molten salt at room temperature. It is interesting to compare ionic motion in ionic liquids with those in superionic conductor.

We have observed millimeter wave absorption bands around 6 and 8 cm⁻¹ in AgI-superionic conductive glasses. These bands were also observed in CuI-superionic ones[1-3]. These bands seems to be due to collective motion of conductive ions because of their frequency, although how conduction ions moves in correlation is not clear.

Recently, we have observed sub-terahertz absorption bands of ILs[4,5]. These bands seems to be due to ionic movement because of their temperature dependence. That is to say, the absorption disappear at low temperatures at which the IL is solid. However, the thermal change of these bands were in various way. This difference seems to be due to the difference of their phase transition to the solid states (crystal or glass).

In this study we have measured millimeter wave absorption spectra of 33 ILs to make clear the ionic movement from the temperature change of absorption spectra.

EXPERIMENTS: A fixed amount of ionic liquids (Tokyo Chemical Industry Co., Ltd.) were spread into filter paper. Transmission spectra of single and double papers with ionic liquids were measured at room temperature and low temperatures. Absorption spectra were obtained by subtraction of them.

RESULTS: Three types of the spectral change occurred at temperature above melting point or glass transition temperature[5]. The absorption bands in the ILs which are crystal in the solid state showed rapid decrease at their melting points[4]. On the other hand, those in ILs which are vitreous in the solid state weakened and shifted gradually. Others showed complicated change as shown in Figs. 1 and 2.

Fig. 1 shows absorption spectra of Triethylsulfonium bis(trifluoromethane)sulfonimide([TES][TFSI]). Figs. 1 and 2 shows increment spectra from that at 88K. At this temperature, these ILs are solid and have only little absorption in this spectral range. At 158K, all four absorption bands decreased below the baseline.

Fig. 2 shows absorption spectra of Tributyl(2-methyloxyethyl)-phosphonium



Fig. 1. Absorption increment spectra of [TES][TFSI]. The baseline is the absorption spectrum at 88 K.



Fig. 2. Absorption increment spectra of [TBMOEP][TFSI]. The baseline is the absorption spectrum at 88 K.

bis(trifluoromethane)sulfonimide([TBMOEP][TFSI]). The 9.5 cm⁻¹ band decreased below the baseline at 158 K.

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Synthesis of Metal Nanoparticles and Nanocomposites by Gamma-ray Irradiation Reduction Method

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INTRODUCTION: Metallic nanoparticles have some specific properties, which are not appeared in bulk materials such as catalytic activities, magnetic properties, electro conductivity and light absorption. These properties depend on its size, shape, structure, chemical composition and so on. They have many possibilities to applied for various industrial fields. Generally, many kinds of metal nanoparticles commercially are synthesized by using chemical reaction method. Recent years, some reports show that it is possible to fabricate various metal nanoparticles under irradiation reduction fields such as ultrasonic, electron beam, ion beam and gamma-ray irradiation. So far, we have reported that synthesis of shape and size controlled Au, Pd, Pt, Cu and these complex nanoparticles by these reaction fields. On the other hand, it is possible that graphene with Pt nanocomposits have high catalytic activity and selectivity due to their excellent properties of graphene and Pt nanoparticles. We have developed a one-step gamma-ray irradiation method to synthesize nanocomposites composed of graphene and Pt nanoparticles from aqueous solution containing graphene and Pt ions in water [1].

EXPERIMENTS: Aqueous solutions with a given concentration of Hydrogen hexachloroplatinate (IV) hexahydrate (H₂PtCl₆6H₂O, Pt(IV)), SDS or 2-propanol. Graphenepowder (6 - 8 nm thick x 5 μ m wide) was added into this water solution. The solution was argon gas purged and sealed into polystyrene vessels. After dispersion by an ultrasonic cleaning bath, they were irradiated at about 300 K with y-rays from ⁶⁰Co radio active source at gamma irradiation facility in KURRI, Kyoto University. The total dose was fixed to 10 kGy with dose rate of 15 kGy/h. After irradiation, the products were separated by centrifugation, washed with water and dried by freeze-drying. UV-vis absorption spectra were measured and all products were observed by conventional TEM. Infrared spectra and Raman spectra (RS) of the prepared samples were collected by a Fourier-transform infrared spectro-photometer (FT-IR)

and a Raman spectrophotometer.

RESULTS:

Figure 1 shows the UV/vis absorption spectra for aqueous solutions before and after gamma-ray irradiation. The absorption peak around 260 nm, which shows Pt complex ions, disappears after y-ray irradiation. This shows that Pt complex ions were reduced by γ -ray induced reductive radicals such as hydrated electron (e_{aq}) and hydrogen radicals. Figure 2 shows the TEM images of Pt and graphene solution sample including SDS and 2-propanol as an additive after y-ray irradiation. Pt nanoparticles homogeneously supported on graphene were formed. The avarage size of Pt nanoparticles was about 30 nm. On the other hand, from the results of FT-IR and RS experiments, it was confirmed that graphene surface was chemically modified by the carbonil groups formed after irradiation. Consequently, fabricated Pt nanoparticles under y-ray irradiation field were supported on the surface modified graphene.



Fig. 1 UV/vis absorption spectra of aqueous solutions of Pt and graphene before and after gamma-ray irradiation.



Fig. 2 TEM images of Pt nanoparticles supported on graphene surface synthesized by gamma-ray irradiation.

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Gamma-ray Induced Photocurrent of Field Emitter Array and Cadmium Telluride-Cadmium Sulfied Photodiode

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INTRODUCTION: Vacuum image sensors with the combination of a field emitter array (FEA) and a photodiode are possible candidate as an elemental part of the camera for observation of interior of the pressure vessel of Fukushima nuclear power plant, the 1st [1]. We have been investigating the deterioration of the performance of FEA and cadmium telluride-cadmium sulfide (CdTe/CdS) photodiode since 2014. These elements already exhibited sufficient endurance against γ -ray irradiation up to 200 kGy. However, the tests have been done after the γ -ray irradiation, and therefore little information on the change in the property under γ -ray irradiation was obtained. There exist many phenomena associated with the γ -ray irradiation, including generation of photocurrent, which prevent us from detecting image signal. This report describes the results of the measurements of photocurrent of FEA and photodiode.

EXPERIMENTS: Six FEAs with different sizes including those of which side length is either 40 or 60 µm were mounted on a single chip. Each FEA possessed a focus electrode in addition to the emitter and the gate [2]. Each FEA has the insulating layer between the emitter and the gate, and also between the gate and the focus electrode. The chip was mounted on the 28-pin LSI package, and was installed in a vacuum vessel. The CdTe and CdS films were prepared by close space sublimation technique and CVD [3]. Gamma-ray irradiation was performed at Gamma-ray Irradiation Facility, Research Reactor Institute, Kyoto University. Current-voltage (I-V) characteristics of the FEAs and the photodiodes were measured during irradiation of γ -ray. The irradiations were done at 20, 30, and 40 cm from the radiation source to check the dose rate dependence. The absorbed doses at 20 and 40 cm were 1.58 and 0.33 kGy h⁻¹, respectively.

RESULTS: Fig. 1 shows the *I-V* characteristics of the photodiode; dashed line, solid line, and dotted line show the dark current, the current during γ -ray irradiation, and the current with visible light at 20 cm apart from the γ -ray source. The photocurrent due to the γ -ray irradiation was almost proportional to the dose rate and as large as 400-500 nA at the bias of -10 V under the absorbed dose rate of 1.58 kGy h⁻¹. Fig. 2 shows the *I-V* character-

istics of the 60 µm-FEA. For the 40 µm-FEA, similar behavior was observed. In this case, both emitter current I_E and gate current I_G were measured individually. The bias was given only to the emitter. Without γ -ray irradiation, both I_E and I_G were almost zero. With γ -ray irradiation, I_E and I_G showed linear increases with applied voltage. However, the I_G did not meet with the I_E . The reason for this has not yet been clarified. Since the irradiation for each condition, no degradation of the samples were observed after the irradiation. Separately performed experiments revealed that the FEA and the photodiodes had sufficient tolerance up to 1.2 MGy.

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Fig. 1. *I-V* characteristics of CdTe/CdS photodiode with and without γ -ray irradiation.



Fig. 2. *I-V* characteristics of FEA under γ -ray irradiation.

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CO4-4

The Development of AMOC System for KUR Positron Beam Facility

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INTRODUCTION: Low-energy antiparticles are becoming increasingly important for both scientific and technological applications. In particular, positrons offer new ways to study a wide range of phenomena including scattering with atoms and molecules, the Fermi surface of metal, and defects and ultrafine precipitates in solids.

The ultrafine precipitation of Cu impurities contained in the old light water reactor pressure vessel (RPV) steels are considered to be one of the main origins for the embrittlement of old RPV. The formation of these ultrafine Cu precipitates is enhanced by the neutron irradiation. This mechanism of embrittlement is one of the topical issues for safety operation of reactors in the field of nuclear industry.

Positron annihilation spectroscopy is one of the powerful tools for detecting sensitively the vacancy type defects in metals and positron affinity precipitates such as the ultrafine Cu precipitates in Fe. In order to investigate the correlation between the formation of the Cu precipitates and the defect clusters induced by neutron irradiation, the technique of the positron age-momentum correlation (AMOC) using high intensity slow positron beam is required. Then, we are constructing a new positron beam facility with high positron intensity at the Kyoto University Research Reactor (KUR), which is the first reactor based positron beam in Japan [1]. The first slow positron beam was successfully obtained from B1-hole at KUR two years ago [2]. Since then, the KUR operation have been stopped. In this work, the AMOC system that conforms to the measurement using KUR positron beam facility was developed during the reactor shutdown.

EXPERIMENTS and RESULTS: AMOC is a technique that allows direct observation of the time dependent transitions of the positron states through simultaneous measurements of both the lifetime and the momentum distributions of annihilating electron-positron pairs. In case of the conventional 3-gamma coincidence system that employ Na-22 for positron source, one nuclear gamma ray of 1.275MeV is used for time determination of positron creation, and two annihilation gamma rays are used for time determination of positron annihilation and measuring momentum distribution of electron-positron pair. In case of the measurements using KUR positron beam, the start signal should be acquired from pulsing trigger of buncher unit instead nuclear gamma ray, which can convert the continuous positron beam from the reactor into a pulsed beam, as shown in Figure 1. The coincidence module is consisted of Fast Amplifier, Timing Filter Amplifier, Constant Fraction Discriminator, Gate and Delay

Generator, and Universal Coincidence etc. Wave shapes from a scintillation detector, a high-purity Ge detector and the pulsing trigger will be directly recorded by the digital oscilloscope with a trigger from the coincidence module. Figure 2 shows a test result of 2D-AMOC spectrum for pure Fe measured by this system. The usual positron lifetime spectrum and the Doppler broadening energy spectrum are obtained by integrating with respect to the momentum and the time, respectively. On the other hand, the time dependent momentum distribution extracted from AMOC spectrum can give the chemical information on the positron trapping site every annihilation times, which is important to understand the correlation between precipitates and vacancy type defects.



Figure 1: Schematic diagram of conventional 3 gamma AMOC; The start signal should be changed to pulsing trigger under the measurement using KUR positron beam.



Figure 2: AMOC 2D spectrum of pure Fe.

We should develop and set up the beam bunching system which is consisted of the chopper, the pre-buncher and the buncher, and the brightness enhancement system which is consisted of a remoderator, some magnetic and electrical lenses. The AMOC measurement will be performed after restarting KUR.

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CO4-5 Basic Study on Radiation-induced Luminescence from Natural Mineral

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INTRODUCTION: It is well known that dielectric materials such as natural quartz exposed to ionizing radiation emit thermoluminescence (TL) and optically stimulated luminescence (OSL). OSL is a well-established tool for measuring radiation doses in unfired sediments [1]. It has features in common with TL which has long been used in measuring radiation doses [2]. Both TL and OSL dosimetry with white mineral do not need to be specially installed in advance, prior to dose estimation.

Quartz is an excellent material for use in dosimetry, because of its almost ubiquitous availability including an accidental place. Feldspar is an extensive ternary family of minerals appropriate for OSL and TL dosimetry as they display a strong luminescence and are quite common in the Earth's formation, although it has anomalous fading effects which decay OSL-related luminescence signals.

However, the detailed emission mechanisms of TL and OSL from natural minerals such as natural quartz and feldspar are not yet clear. In this study, the emission mechanisms of TL and OSL were investigated in conjunction with various radiation-induced phenomena after annealing treatments of quartz samples, involving TL, OSL and electron spin resonance (ESR) measurements.

EXPERIMENTS: Two surface soil samples were collected at different places in Ibaraki. Natural quartz samples were extracted from the soils by a usual treatment of 6M hydrochloric acid (HCl), 6M sodium hydroxide (NaOH) followed by 60 min of concentrated hydrofluoric acid (HF). The etching treatment with HF solution was at room temperature. Further purification of the quartz grains was performed by hand selection for the sake of elimination of feldspar grains as low as possible under a fluorescent lamp. The purified quartz samples were sieved to adjust the grain sizes ranging from 150 µm to 250 µm in a diameter for ESR spectrometry. The quartz samples were annealed at 800 °C for 24 hours in an electric furnace, to make luminescence signals strong. The annealed quartz samples were irradiated a dose of 1 kGy with ⁶⁰Co source (0.64 kGy/h at D-30 cm) at room temperature at Kyoto University Research Reactor Institute (KURRI). The irradiated samples were stored at room

temperature for one day to eliminate afterglow emission in a dark room. The ESR measurement was carried out using an ESR spectrometer (Jeol Ltd., JES-TE 200) at -196 °C. Prior to the ESR measurements, the quartz samples were annealed for 1 min at 50 °C intervals ranging from 150 to 300 °C with a ceramic heater as preheat treatment. A part of each sample from two sampling points was kept in a dark room without preheating treatment. All preparations were carried out under dim red light.

After the ESR measurements, all samples were kept to measure luminescence signals in a dark room. However, an automated TL/OSL-reader system (JREC) had been improved to heat sample at correct temperature and then could not measure TL and OSL signals.

In this research, there were our two chances to do ESR experiment at the KURRI, but one of the chances was lost to measure ESR signals because the ESR spectrometer could not be worked in last August.

RESULTS: The ESR signals of Al centers as hole-trapped centers and Ti centers ($[TiO_4/H^+]^0$, $[TiO_4/Li^+]^0$ and $[TiO_4/Na^+]^0$) as electron-trapped centers were detected in the annealed quartz samples as before. ESR signal intensity of Al centers decreased with preheating temperatures, gradually. Finally, the ESR signals at 300 °C of preheat were almost disappeared.

On the other hand, ESR signal intensity of Ti centers showed different tendency from the ESR intensity of Al centers. The intensity of Ti centers was constant up to 150 °C of preheating temperature. After heating over 150 °C, the intensity of Ti center decreased with preheating temperatures. Finally, the ESR signals at 300 °C of preheat were almost disappeared as well as Al centers. The results agreed to our previous results. However, the intensity tendency of Ti centers was different between two quartz samples.

In this research, the luminescence emission mechanism could not be identified. Therefore, further work is necessary to identify luminescence mechanism using ESR measurement and annealing experiment. Moreover, the sample measured by ESR spectrometer should be used in luminescence measurement.

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CO4-6 Study of the Scientific Properties of Water and the Physiological Activation Phenomena by Using High-Intensity Pulsed Coherent Radiation

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INTRODUCTION: The coherent transition radiation (CTR) from electron bunches of a linear accelerator (linac) has continuous spectra in a submillimeter to millimeter wavelength range. It is a picosecond pulsed light and hence, has extremely high peak-intensities compared with the other terahertz (THz) light sources. The light sources using the CTR have been applied to absorption spectroscopy [1-4]. However, such light sources are very limited. The light source system using the CTR from the electron beams of the 45 MeV L-band electron linac has been established at KURRI [5]. The CTR light source developed has been applied to absorption spectroscopy and it is expected that any nonlinear effects might be observed. Another important application is to the investigation of the biological effects of the CTR; few experiment has been performed so far.

The main purpose of the present work is the investigation of the biological effects of the high-intensity pulsed CTR.

CHARACTERISTICS OF THE CTR: The CTR is emitted as a pulsed and polarized radiation. The micropulse structure of the CTR corresponds to that of the electron beam from the linac. The light can produce the intense pulsed electric field and would cause some kind of excitation in a matter. The spectrum of the CTR is determined by the micropulse shape of the electron bunch whose pulse width is usually about 10 ps in the case of an L-band linac.



Fig. 1. Schematic diagram showing the configurations for absorption spectroscopy using the CTR

EXPERIMENTAL METHOD: The experimental configurations for the absorption spectroscopy are schematically shown in Fig. 1. The output CTR light from a light source chamber was transported out from the accelerator room. The spectrum of the light after passing through the sample was measured with a Martin-Puplett type interferometer and a liquid-He-cooled silicon bolometer. The sample was located on the light path between the interferometer and the detector. The light was focused at a light collimator 8 mm ϕ in diameter located before the sample. The details of the methods for the measurements are described in ref. 5.

The liquid sam-

ples used in the experiments are water, aqueous solutions of NaCl, several kinds of bacillus, cell and microorganism. In the irradiation ex-



periments of the CTR the sample holder shown in Fig. 2 was located just before the interfer-



ometer on the light path. The thickness of the liquid sample was about 100 μ m, which was sandwiched with two anhydrous quartz plates 3 mm thick.

RESULTS AND DISCUSSION: The basic properties of the CTR light source were investigated [6]. The light spectrum was sufficiently stable during the measurements within ± 2 -3% in a wavenumber range of 4-13 cm⁻¹. The range can be expanded to the lower or the higher wave numbers by using light filters. The wavenumber resolution was 0.1 cm⁻¹. The intensity of light was found to be sufficiently high even if it becomes 10⁻⁶ of the initial one after transmission through the sample due to absorption.

Some preliminary experiments to optimize the experimental conditions have been performed so far.

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CO4-7 Analysis of Circularly Polarized CTR Using Babinet-Soleil Compensator

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INTRODUCTION: In recent years various types of coherent radiation emitted from a short bunch of relativistic electrons have attracted a considerable attention as a bright light source in the THz-wave and millimeter wave regions for the spectroscopic purpose. Coherent transition radiation (CTR), which is emitted from a boundary between two media, is one of such a coherent light source. CTR is usually utilized as a non-polarized light source, because the electric vector of transition radiation (TR) emitted from a metallic screen is axially symmetric with respect to the trajectory of an electron beam. In my previous reports [1] the circularly polarized CTR using a pair of wire-grid radiators with the different polarization has been developed with a new idea. Shibata et al. has developed a technique of generation of circularly polarized millimeter-wave radiation with the phase difference between the forward TR and the backward one [2]. However, it was difficult to control the polarization degree in that technique. The significant point of my new technique is the use of linearly polarized CTR with the wire-grid radiator. With this technique the polarization degree is able to be controlled precisely. In this report circularly polarized CTR has been analyzed a hand-made Babinet-soleil compensator. using Circularly polarized light has been useful in the circular dichroism spectroscopy.

EXPERIMENTAL PROCEDURES: The experiment was performed at the coherent radiation beamline [3] at the L-band linac of the Research Reactor Institute, Kyoto University. The energy, the width of the macro pulse, and the repetition rate of the electron beam were 42 MeV, 33 ns, and 20 Hz, respectively. The average current of the electron beam was 0.5 µA. The schematic diagram around the radiation source of this experiment is shown in Fig.1. Two wire-grid polarizers (WG1 and WG2) 10 µm thick with 25 µm spacing were used in order to generate the CTR with horizontal and vertical components, respectively. Each CTR was superposed with phase difference through the optical delay system. Although a Martin-Puplett-type interferometer is usually used as a spectrometer, a grating-type monochromator was used in this experiment. The monochromator was set for the wavelength of 2.3 mm. The CTR was detected by a liquid-helium-cooled Si bolometer. In order to measure the polarization diagram a wire-grid polarizer with a rotary holder and a Babinet-soleil compensator were used

in front of the detector.

RESULTS: The observed polarization diagrams of circularly polarized CTR at the phase difference of $\pi/2$ and $3\pi/2$ are shown in the left figures in Figs.1 and 2, respectively. In order to analyze the circular polarization the thickness of the Babinet-soleil compensator was set as a quarter-wave retarder. The observed light through the compensator has not perfect linear polarization as shown in the right figures in Figs.1 and 2. These results have been attributed to the lack of coherence of CTR because of the large size of the electron beam.

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Fig.1. The schematic diagram of the experiment.



Fig.2. Circularly polarized CTR at the phase difference $\pi/2$ (left) and polarization diagram through the compensator (right).



Fig.3. Circularly polarized CTR at the phase difference $3\pi/2$ (left) and polarization diagram through the compensator (right).

CO4-8

⁵⁷Fe Mössbauer Spectra of Normalconducting Sr₂CrFeAsO_{3-d}

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INTRODUCTION: Layered oxypnictide $Ae_2T_MFePnO_{3-d}$ (Ae = Alkaline earth metal, TM = Transition metal) (21113) consists of carrier conducting FePn (Pn = P and As) layer and carrier blocking $Ae_2T_MO_3$ layer. Several 21113 shows superconducting phase at low temperatures. Hyperfine fields (B_{hf}) measurements experienced by nuclei of atoms at crystallographic sites in magnetic and/or antiferromagnetic metals are of interest in view point of hyperfine interactions and important for their use in the determinations of nuclear magnetic dipole moments by a variety of methods including those based on low-temperature nuclear orientation. Guang-Han Cao et al reported that Sr₂VFeAsO_{3-d} is an FeAs-based layered oxypnictide superconductor, which reveals antiferromagnetic under 150 K originating in local d-electron moments of vanadium, and the superconducting transition temperature T_c is 24 K.[1] In this study, we focus on magnetic phase of Sr₂CrFeAsO_{3-d} as a possible mother compound of iron-based superconductors. [1-5]

Element-specific magnetism and electronic phase of Fe sublattice in Sr₂CrFeAsO_{3-d} are elucidated using ⁵⁷Fe Mössbauer spectroscopy. Studies on hyperfine fields B_{hf} experienced by nuclei of impurity phases are of interest in view point of hyperfine interactions and important for their use in the determinations of nuclear magnetic dipole moments by a variety of methods including those based on low-temperature nuclear orientation.

EXPERIMENTS: Polycrystalline samples of Sr₂CrFeAsO_{3-d} were prepared by the solid-state reactions in a sealed silica tube using dehydrated SrO, FeAs, Cr₂O₃, Cr as starting materials. Then, a mixture of the three powders was pressed into pellet and heated in doubly sealed silica tubes at 1030~1330 °C for 40 h. All procedures were carried out in an Ar-filled glove box (MIWA Mfg; O_2 , $H_2O < 1$ ppm). The crystal phase of the obtained product was examined by powder X-ray diffraction (XRD; RigakuRINT-2500) using CuKa radiation. Almost all the diffraction peaks are assigned to the Sr₂CrFeAsO_{3-d} phase. Electrical resistivity measurements were performed at 2-300 K by a dc four-probe technique using silver paste as electrodes. Magnetization measurements were performed for polycrystalline samples using a Quantum Design magnetic properties measurement system (MPMS). The ⁵⁷Fe Mössbauer (MS) experiment on the two series of compounds, $Sr_2CrFeAsO_{3-d}$ (x = 0.03, 0.18) was performed with conventional ⁵⁷Fe MS equipment using 14.4 keV γ -rays from a ⁵⁷Co source in an Rh matrix.

RESULTS: ⁵⁷Fe MS to the samples of d=0.03, 0.18 of Sr₂CrFeAsO_{3-d} were measured and analyzed for obtaining quantitative value of internal magnetic field. As a result,

the existence of spin density wave phase are observed for x = 0.18, while antiferromagnetic ordered Fe are observed for x = 0.03. The result indicates that normalconducting Sr₂CrFeAsO_{3-d} shows two magnetic ground states, which stabilities depend on oxygen deficiency.



Fig. 1. ⁵⁷Fe Mössbauer spectra (MS) of polycrystalline $Sr_2CrFeAsO_{3-d}$ (d = 0.03 and 0.18) at several temperatures described in the figure. The solid lines for d = 0.03 are fitted patterns provided that a internal magnetic field. The solid lines for d = 0.18 are fitted patterns provided that a wide distribution of internal magnetic field. Profiles of FeAs, which are denoted by Green lines, are reproduced from Blachowski et al's results. [6,7]

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CO4-9

Complex Structure of Ions Coordinated with Hydrophilic Polymer. 16. Application for Metaric Plating on Resin Surface. (1)

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INTRODUCTION: We have reported interacted structures between iodine (polyiodide ions, I_n^{m-} , *m*, *n*: integer, $n \neq 1$) and polymers.[1] While polyiodide ions (and their counter-ions) are prepared as solutes in aqueous solution, they can be diffused into various polymeric matrix. Such structures and procedures are expected to introduce novel functionality and wide applications to polymeric materials with easy operation.[2]

For some hydrophilic polymers, diffusion of iodine from aqueous solutions advances very quickly and, in some matrix such as polyamide-6 (PA6), ionic diffusion into crystallite region or "diffusion-induced" orientation are also observed through easy operation at room temperature; as phenomena, iodine-doped polymers can be regarded as "pseudo solvents" for ionic diffusion.[3,4]

These results suggest that coordination between iodine and polymers is dynamic and pliable and activated process and that previous existence of polyiodide ion drastically enhances posterior ionic diffusion in polymers. On the other hand, self-organization and diffusion keeping ordered structure are also observed; processes looked opposite or paradoxical go simultaneously as both ordering and disordering effects. Furthermore, such ionic behavior can be qualitatively applicable for <u>hydrophobic</u> matrix even if sample operation are wholly carried out with <u>aqueous</u> solution of polyiodide ions.[5]

Some advantages can be suggested as availability for such operation using polyiodide ions: maintenance of shape or convenience of size of matrix, deep and gradient injection of inorganic components in spite of operation even at room temperature, preparation with aqueous solutions,,, etc. Previously, we reported "(1st) iodine doping" and "secondary doping" of Ag^+ ion into hydrophobic materials with aqueous solutions.[6] Here, injection of metallic ions or inorganic salts into polymeric matrix are expected to apply electroless plating resin surface.

EXPERIMENTS: As hydrophobic materials as matrix, silicone-rubber tube (LABORAN, $\phi 2x4$), PE (polyethylene) tube and PP (polypropylene) film were treated. These matrixes were "iodine doped" by immersing in I₂-KI(aq) or I₂-NH₄I(aq) (3.0N) for more than one week. For secondary doping of Ag⁺ ion, each samples were immersed in AgNO₃(aq) (2.0M) solution for a few days. On electroless plating of Cu in aqueous solution, THRU-CUP PSY (UYEMURA) was used. All opera-tion were done at room temperature (c.a. 25°C).

RESULTS: Intrinsically, the treated three matrixes are

hydrophobic as chemical composition. Actually, affinity with each aqueous solutions was not so activated in each process without "iodine doping". And, even applying "iodine doping", increase in mass (Δm) in each preparation steps were tiny (less than 1-2%) and colored appearance on "(1st) iodine doping" was negligibly slight while, to hydrophilic matrixes, $\Delta m = 50-200\%$ (or more) or solid and tough "black" coloring were frequently observed for "iodine doping".[1] However, as results after electroless Cu plating process to the hydrophobic matrixes, deposition of Cu was observed on surface of each hydrophobic matrixes.(Figure)

Such Cu deposition is regarded as process at restricted surface region of each matrixes. In the cases of hydrophobic matrixes, diffusion of polyiodide ions might be qualitatively very slight and following ionic diffusion introduced posteriorly is also negligible; ionic diffusion and exchange (ex. polyiodide ions, counter ions, "secondarily doped" ions,,,) are hardly explicit like as hydrophilic systems. Nevertheless, results showing activated affinity with aqueous solutions means that "iodine doping" can suggest potential abundance for functionality to

unexpected matrixes.

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Figure: Electoless plating for hydrophobic matrixes treated with aqueous solutions for all process after applying polyiodide: silicone rubber tube (top), PE tube (center), PP film (bottom).

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CO4-10 Interactions of Deuterium with Defects Created in Tungsten by Irradiation of High Energy Electrons

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INTRODUCTION: Retention of hydrogen isotopes especially tritium in plasma facing materials is one of the important issues in safety assessment of fusion reactors. Recent report shows that neutron irradiation significantly increases deuterium (D) retention in W, a leading candidate of plasma-facing material, due to trapping effects of radiation-induced defects [1]. However, detailed trapping mechanisms have not been clarified. For better understanding of defect-hydrogen isotope interactions, in this study, W specimens were irradiated with high energy electrons to induce Frenkel pairs uniformly throughout the bulk, and clustering of vacancy-type defects and D trapping were examined using positron annihilation spectroscopy (PAS) and thermal desorption spectroscopy (TDS).

EXPERIMENTS: Disks of recrystallized W (RC-W) and stress-relieved W (SR-W) were irradiated with 8.5 MeV electrons at around 373 K to 10^{-3} dpa. Then those specimens were subjected to exposure to D₂ gas (0.1 MPa) or annealing in vacuum at 573 K for 100 h. Size distributions of vacancy-type defects were examined using PAS, and D retention was measured using TDS.

RESULTS AND DISCUSSION: The results of positron lifetime measurements for RC-W are summarized in Table 1. The positron lifetime before the irradiation was 118 ps. It clearly increased to 170 ps by the electron irradiation due to formation of dislocations, mono- and/or divacancies. Long lifetime components appeared after D₂ gas exposure and annealing in vacuum at 573 K, and the positron lifetime after D_2 gas exposure (~280 ps) was significantly shorter than that after annealing in vacuum (~342 ps). The increase in positron lifetime showed the clustering of vacancies. The difference between D₂ gas exposure and annealing in vacuum can be explained by retardation of clustering of vacancies by D in W and/or decrease in the effective open-volume in vacancy clusters due to D trapping. Similar change in the lifetime was observed also for SR-W. Coincidence Doppler broadening spectroscopy showed that the correlations between S and W parameters for the specimens before irradiation, after irradiation and after annealing in vacuum fell on a single straight line in S-W plot, while the specimen exposed to D₂ gas after the electron irradiation showed the TDS showed significant increase in D retention due to the irradiation, as shown in Fig. 2; D concentration in RC-W increased from $[D]/[W] = 1.1 \times 10^{-6}$ to $[D]/[W] = 8.6 \times 10^{-6}$. A desorption peak appeared at 850 K and a shoulder was observed at 975 K. These two desorption stages appeared to correspond to the detrapping of D from small and large vacancy clusters observed using PAS. The evaluation using a diffusion analysis program showed that these desorption temperatures correspond to the activation energy for detrapping of 1.55 and 1.85 eV. Interestingly, D retention in the irradiated SR-W was larger than that in the irradiated RC-W and it was $[D]/[W] = 5.5 \times 10^{-5}$.

Table 1 Positron lifetime in W under as-prepared conditions (non-irr.), after electron irradiation at ~373 K to 10^{-3} dpa (e-irr.) and subsequent annealing at 573 K for 100 h in vacuum (e-irr. vac. anneal.) and in 0.1 MPa D₂ gas (e-irr. D₂ exp.).

Sample		non-irr.	e-irr.	e-irr. vac. anneal.	e-irr. D ₂ exp.
Positron life time (ps)	Average	118	170	231	194
	Short life compo- nent	-	-	130	134
	Long life compo- nent	-	-	342	280



Fig. 2 Thermal desorption spectra of D from the recrystallized W specimen under as-prepared conditions and that irradiated with 8.5 MeV electrons to 10^{-3} dpa.

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CO4-11 Effect of Ion Species on Defect Formation near Surface of GaSb by Ion Irradiaiton

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INTRODUCTION: Porous structure is formed on GaSb surface by ion beam irradiation [1], [2]. It was clarified that the structure is formed by migration of point defects induced by ion irradiation. The first the voids form from the oversaturated vacancies and they develop by movement of vacancies and interstitials. However, the the detailed process of void formation has not yet been understood. In this study, the effect of various ion species on GaSb void formation by ion irradiation was investigated. The ion species used in this work are ions of carbon group elements, by which it will be possible to detect only the effect of ion mass except the chemical effect.

EXPERIMENTS: GaSb (001) wafers were irradiated at about 100 K by C, Si, Ge, Sn, Pb using a heavy ion accelerator at Research Reactor Institute, Kyoto University. The acceleration voltage was 60 kV. The ion dose was 2.80×10^{19} ions/m² for C, 1.57×10^{19} ions/m² for Si, 1.01×10^{19} ions/m² for Ge, 6.02×10^{18} ions/m² for Sn, and 7.05×10^{18} ions/m² for Pb. These ion doses were determined to gives a same peak concentration in depth profile of vacancy concentration in GaSb calculated by SRIM code simulation [3], adopting the displacement threshold energy values obtained by Thommen (6.2 eV for Ga and 7.5 eV for Sb) [4]. The surface morphology was observed by a FE-SEM (field emission scanning electron microscope, JEOL JSM-7401F).

RESULTS AND DISCUSSION: The voids were observed on the surfaces irradiated by relatively large mass ions (Ge, Sn, and Pb), and they were scarcely formed on the surfaces irradiated by the small mass ions (C and Si). Figure 1 shows distributions of vacancy created by one ion irradiation obtained by SRIM code simulation. The distribution of point defects in depth direction is much different between ion species. In the case of light ions, vacancies are formed in deep region from the surface and the local vacancy concentration is low. In the case of heavy ion, the vacancies are created in shallow region and its local concentration is high, when they aggregate easily and lead to void formation. In contrast, vacancies created at low concentration in wider region diffuse so that void formation scarcely occurs. This is the explanation that the surface structures were different between heavy ion irradiation (Ge, Sn and Pb) and light ion irradiation (C and Si).



Fig. 1. Vacancy distributions of one ion irradiation obtained by SRIM code simulation.

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INTRODUCTION: The Ganymede Laser Altimeter (GALA) as part of the JUICE (Jovian Icy Satellite Explorer) payload is one of the instruments focusing on aspects related to the presence and characterizations of subsurface water oceans [1] [2]. For the first time the time-variability of the global figure of a moon due to tides exerted by Jupiter will be detected by altimetry measurements.

Laser pulses are emitted at a wavelength of 1064 nm by using a Q-switched Nd:Yag laser at operational frequencies of 30 and 75 Hz, respectively. The pulse energy and pulse repetition frequency depend on the different JUICE mission phases. The beam is reflected from the surface (surface spot size is about 50 m) and received around 3 ms (assuming the 500 km orbit) later at a 25 cm diameter F/1 telescope. The returning laser pulse is refocused onto a silicon avalanche photodiode (or APD) through a narrow bandpass interference filter. The signal is then sampled and fed to a digital range finder. This system determines the time of flight (and therefore range), the integrated pulse intensity, its width and full shape. The data are passed to a digital processing unit which controls the operation and services the spacecraft interface. GALA will provide <1 ns time resolution (<15 cm range, <8 cm under optimal conditions).

APD for GALA: We adopted an APD product manufactured by Excelitas Technologies, Montreal in Canada. They commercially manufacture APD product particularly a hybrid module of APD which contains an APD sensor, a preamplifier, a thermo-electric controller and a temperature sensor in a package. Their APD product that are customized for space use have been adopted for space missions due to its compactness and heritage in space.

The APD has also sensitivity to radiation that induces hole-electron pair in the depression layer. In space environment, the induced signal may affect light detection as background noise, called radiation noise. Jupiter has a strong magnetosphere and previous studies revealed energetic electrons are trapped in the magnetosphere and major component among the other ionizing radiation like proton and the other energetic ion. The APD will be shielded up against to the Jovian radiation environment however some of electrons can penetrate the shield into the APD. In this study, we used KURRI-LINAC as an electron beam source to emulate electrons irradiating the APD to investigate how energetic electron induces signals and interferes the returning laser pulse signal. **EXPERIMENTS:** We used an APD product of C30954E, Excelitas Technologies, which is the same product as one for GALA. The APD sensor has a diameter of 0.8 mm and an enhanced sensitivity in infrared range, about 0.4 of quantum efficiency at 1064 nm. It does not include any preamplifier so a preamplifier was prepared and externally attached to the device. The amplifier is a trans-impedance amplifier, the same as one for GALA flight APD. Although the band width for GALA APD had not been decided yet, 100 MHz was adopted as plausible number for the GALA APD.

In an irradiation test, 5 MeV electron were irradiated to the APD. During irradiation, reverse bias high voltage was applied to the APD sensor and the ground line was fed to the following preamplifier. The output signals of the amplifier were monitored and recorded with a digital oscilloscope.

RESULTS: As shown in Fig. 1, clear pulse signals were detected and they correspond to vertical hits of single electron on the APD sensor. In actual environment, energetic electron with various energies omni-directionaly comes and penetrates the sensor. And also the APD sensor will be sit in the radiation shield where secondary radiation are induced affecting the optical returning pulses. Those will be investigated in further experiments.



Fig.1. An example of output signal pulse from the APD.

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CO5-1

Size Distribution and Concentration Change of Antimony in the Atmospheric Aerosols at Sakai, Osaka

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Antimony is a metal element, of which symbol Sb; atomic number 51;atomic weight 121.76;melting point 630.5°C. It is used in alloys with lead in battery plates and flame-proofing compound. Antimony(Sb) is toxic by small amount (100mg) into body. As it exist naturally in the soil we get it as food contamination from which Sb is absorbed into the body by 0.002-1.mg per day. Also the airborne particulate matter, aerosol in the atmosphere contain antimony as the fume from burning of the waste matter. This suggest that the daily inhaling of Sb from ambient atmosphere could influence our health. To estimate the effect of Sb to our health ,here we report the size distribution and the concentration(mean and variation) and monthly concentration change.

The analyzed samples are size-separated aerosols and non size-separated aerosol. The size-separated samples have been collected with 9-size particle size ranges in 1 weak sampling period. The non size-separated samples have been collected daily on membrane filter in 24h sampling period. The sampler of both samples are located at Osaka Prefecture University, at Sakai.

The element of the samples were analyzed by neutron activation analysis using KUR. The Analysis condition for Sb are , Irradiation : 2 hours in Pn2 1MW, detection of gamma ray : 124 Sb 1661kev.

From the analyzed results the size distribution of Sb in atmospheric aerosol can been seen as mostly concentration distribute in very fine range , <0.43 μ m(Fig.1). The central diameter and concentrations of samples collected in 2012 are described on table 1.

Table 1 Central diameter and total concentration of Sbcollected by 9 particle size separation at Sakai in 2012.Pariod(2012)D50(mm)TC(n a(m2))

Period(2012)	D50(µm)	1C(ng/m3)
Feb 14-21	0.94	1.7
Apr 3-10	0.68	1.7
Jun 5-12	0.64	1.3
Aug 7-14	0.69	1.2
Oct 9-16	0.67	2.3
Dec 11-18	0.57	3.3



From the result of concentration change in daily collected(in 2007) samples we have obtained concentration frequency distribution(Fig.2), showing mostly frequent in range, 1-3ng/m3. And we have got the



Fig.1 Size distribution of Sb in aerosols collected at Sakai,0214-0221 in 2012.

average and standard deviation, $2.8 \pm 1.9 \text{ ng/m}^3$, min=0.5 ng/m³, max=24 ng/m³. This value indicate that mean inhalation of Sb per day is about 0.06 mg.

Monthly changes(mean, sdev., min.,max.) of daily concentration are shown Fig.3 . Months in high concentration are Jan.(3.4ng/m³), Oct.(3.5ng/m³), Nov. (3.6ng/m³) and lowest on May(2.0ng/m³). Maximum concentration of daily change in 2007 were observed on September 30th. On this day we observed high concentration in bromine(Br), suggesting the source of fume in high concentration might be waste burning of plastic that contain flame-proofing compound.



Fig.2 Frequency distribution for daily concentrations of Sb in arosols collected at Sakai,2007.



Fig.3 Monthly changeof Sb in aerosols collected at Sakai, 2007.

CO5-2

Characteristics of Synthetic Calcite Thermoluminescence Studied for Paleoenvironmental Reconstruction of East Asia

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INTRODUCTION: Luminescence dating observes the natural accumulated radiation damage caused by radioisotopes such as U and Th as the form of glow after stimulation by heating or lightening. Thermally stimulated luminescence from calcite shows strong red emission. However, thermoluminescence dating of calcite is less popular because of some unknown problems; e.g., sensitivity change of calcite occurred through repeated heating of samples, possible anomalous fading, difference in characteristics of luminescence response against different kinds of radiation (e.g., gamma-ray, beta-ray, alpha-ray, and X-ray). When thermoluminescence characteristics of calcites are examined using natural occurring calcite, their response to the various radiations depends on minor chemistry (Fe, Mg, Mn and Sr). In this study, calcites with controlled impurity concentrations were synthesized and analyzed to evaluate relationship between multiple impurity concentration and thermoluminescence properties quantitatively, together with natural calcites.

EXPERIMENTS: Calcite were synthesized from sodium carbonate (>99.8wt%) and calcium chloride (>95 wt%). Iron (II) chloride (>99.0-102.0 wt%), magnesium chloride (>98.0wt%), and manganese chloride (>99.0wt%) were added at different levels to produce calcites with different amount of impurity. Chemical composition of resultant calcite were measured by LA-ICP-MS, and the results were reported in KURRI progress report 2014. Gamma irradiation was carried out at the ⁶⁰Co gamma irradiation facility at Kyoto University Research Reactor. Alpha and beta ray irradiations were carried out with disc-type ²⁴¹Am and ⁹⁰Sr source, respectively. Given doses by Gamma and beta irradiation were estimated by quartz irradiated together with synthesized calcite. A given dose by alpha irradiation was estimated by alpha track counting. Then luminescence emission from calcite was measured by the luminescence reader MOSL-22, and dose was estimated by the external x-ray source, whose dose rate is calibrated as 0.1 Gy/sec with quartz. This measured dose on calcite was compared to the given dose on quartz, and the ratio (calcite/quartz) was calculated for each irradiation (gamma, beta). Results on alpha irradiation will be reported later.

RESULTS: Calcite/quart ratios in dose estimate by luminescence were different among samples. However, ratios by beta-ray and by gamma- rays are more or less concordant (Fig. 1). When Calcite/quartz ratios by gamma dose is plotted against Mn concentration, they show negative correlation (Fig. 2). Thermoluminescence characteristics of calcite is controlled by the Mn impurity. Therefore, measurement of Mn concentration is inevitable for calcite thermoluminescence dating.



Fig. 1. Relationship between calcite/quartz ratios by beta and gamma irradiation.



Fig. 2. Calcite/quartz ratios by gamma irradiation are plotted against Mn concentration.

Study of Earth and Planetary Matters by Thermoluminescence

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INTRODUCTION: Induced TL (thermoluminescence), the response of a luminescent phosphor to a laboratory dose of radiation, reflects the mineralogy and structure of the phosphor, and provides valuable information on the metamorphic and thermal history of meteorites. Thermal metamorphism causes the production of feldspar, the major TL phosphor in ordinary chondrites, by the devitrification of feldspathic mesostasis. TL sensitivity normalized by Dhajala chondrite was measured at around 150 °C or 80 °C in the induced TL intensity. Primitive chondrites of petrologic subtype ≤ 3.4 with coefficient of varia-tions (σ as a percentage of the mean) over 50%, of favalite in the olivine have low TL sensitivities under 0.1. The TL sensitivity was usually used to determine petrologic subtype of unequilibrated ordinary chondrites [1]. On the other hand, the TL sensitivity decreases 10-fold after shock-loading to 25-32 GPa [2], and terrestrial weathering makes the TL sensitivity decrease 16-fold at maximum in Antarctic chondrites [3]. Then these secondary altered chondrites were reported to have low TL sensitivities, equivalent to primitive chondrites [4]. Until now, the induced TL of ordinary chondrites has been measured above room temperature. Last fiscal year, we preliminarily measured low temperature TL from liquid nitrogen temperature of twenty-one Asuka ordinary chondrites under 0.1 in TL sensitivities. This time we measured low temperature TL of additive two Asuka ordinary chondrites, A881397 (LL3.3) and A881236 (remarkably shocked). Also this time, we also measured induced TL of unshocked and artificially shocked terrestrial feldspars, Minas Genas albite, Shiga albite and Niigata plagioclase, to investigate the shock effects. This year, there is no request from NIPR to determine subtype because samples, judged to be unequilibrated, became a little. Then we didn't try to measure new chondrites.



Fig.1. induced TL glow curves of A881397 (LL3.3) and a remarkable shocked chondrite of A881236 (H).

RESULTS: Fig.1 shows TL glow curves of a primitive ordinary chondrite of A881397 (LL3.3) and remarkable shocked chondrite of A881236 (H). There were also two peaks around -120 and -80°C lower than room temperature. However, we could not find the qualitative tendency in induced TL glow curves in all twenty-three chondrites.

Induced TL of terrestrial feldspars, Minas Genas albite, Shiga albite and Niigata plagioclase, showed very high intensity. Shocked samples showed relatively less intensity than unshocked samples. However, we unfortunately couldn't find clear difference in the induced TL glow curves between the unshocked and shocked terrestrial feldspars. Fig.2 shows induced TL glow curves of unshocked and artificially shocked Shiga albite.



Fig.2. Induced TL of unshocked and artificially shocked Shiga albite.

ACKNOWLEGEMENTS: Over twenty years, we have measured induced TL of 407 Antarctic unequilibrated ordinary chondrites, Yamato 242 chondrites (LL3: 30, L3: 113, H3: 99), Asuka 163 chondrites (LL3: 39, L3: 48, H3: 76) and ALH 2 chondrites (L2:, 2), which TL analysis has been requested from NIPR, under the Visiting Researcher's Program of the Research Reactor Institute, Kyoto University We have found 50 samples to be primitive chondrite under ≤ 3.4 in them. This time we finished to apply to this program. We would like to thank to not only staffs in the Research Reactor Institute, who has supported us, but also all staffs in the Research Reactor Reactor Reactor Institute, Kyoto University.

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INTRODUCTION: Characteristics of tritium water (HTO) are almost the same as water (H₂O) and deuterium water (D₂O or HDO). The slight difference of the characteristics (e.g. the boiling temperature and electrolysis etc) have been used to fraction three waters. It was recently reported that manganese oxide adsorbed T^+ [1]. However, it is still difficult to fraction the three waters.

Magnesium is one of the main elements composed of the earth. Magnesium oxide (MgO) easily reacts with water molecules and magnesium hydroxide (Mg(OH)₂) is produced, immediately. Additionally, if MgO is placed in the air, MgO will react with water molecules and become $Mg(OH)_2$ [2]. The mineral name of $Mg(OH)_2$ is brucite. Brucite is produced by weathering of a mafic rock with other clay minerals on the earth's surface. $Mg(OH)_2$ was dehydrated at about 300 °C [3]. Recently, there are some reports about the dehydration of $Mg(OD)_2$ by the X-ray or the neutron diffraction with the heating system. The temperature of the dehydration was about 330 °C [4, 5]. Although the temperature is not certain, the dehydration temperature of Mg(OD)₂ was higher than that of Mg(OH)₂. When MgO reacts with HTO, the reaction rate and dehydration temperature of Mg(OT)₂ might be different from those of Mg(OH)₂.

We investigated that the reaction rate of $Mg(OT)_2$ is different from that of $Mg(OH)_2$, as a first step.

EXPERIMENTS: We used MgO (light and heavy) to be on the market. MgO (light) and (heavy) means a fine particle and a coarse particle, respectively. We prepared six glass bottles with stopper. In every three bottles, 1 g of MgO (light) and 2g of MgO (heavy) were put, respectively and 10 cm⁻³ of water with HTO was added in six bottles. These bottles are left for about 1, 4 or 24 h. After the purpose reaction time, suspension was filtered by 0.2 µm of a disposable filter. We diluted 0.1 cm⁻³ of supernatant by 20 cm⁻³ of the liquid scintillator. The tritium concentration of each sample was measured by a scintillation detector (Packard, Liquid Scintillation Analyzer).

RESULTS: When the tritium concentration of supernatant is smaller than that of the initial solution, more amount of HTO reacts with MgO than that of H_2O . If the tritium concentration of supernatant is larger than that of the initial solution, H_2O will be preferred to HTO in the reaction with MgO. In the case of the similar tritium concentration of the initial solution and supernatant, almost the same amounts of HTO and H_2O react with MgO.

The tritium concentration of supernatant is shown in Table 1. For MgO (light), the tritium concentration decreased for 1 and 4 h after MgO contacted with water with HTO. The removal ratio was about 5.1%. After 24 h, the tritium concentration of supernatant was almost the same as that of the initial solution. The removal ratio was about 1.5%. If MgO contacts with water with HTO for a few hours, the reaction of HTO with MgO might be quicker than that of H₂O. MgO might react with slightly more HTO than H₂O in water for 4 h. After the synthesis of brucite, the exchange reaction would occur between T⁺ or H⁺ in brucite and H⁺ or T⁺ in water. The exchange reaction might achieve the equilibrium for 24 h. The tritium concentration of supernatant after 24 h might be similar to that of the initial solution

For MgO (heavy), we used 2g of MgO, because it was expected that the reaction of MgO and water with HTO would be slow. After 4 h contact, the tritium concentration of supernatant was a smaller than that of the initial solution. The ratio of 2.4 % of T^+ decreased from the solution. This removal ratio was smaller than that of MgO (light). Since MgO (heavy) is a coarse particle, MgO (heavy) would not sufficiently react with water with HTO. The removal ratio of MgO (heavy) would be smaller than that of MgO (light). In 24 h, the removal ratio was about 1.9 %. The tritium concentration of supernatant in MgO (heavy) and the initial solution is almost the same in the case of MgO (light). Probably, T^+ or H^+ in brucite will be exchanged for H^+ or T^+ in water.

Table 1 Change of the tritium concentration in supernatant by the contact time of MgO and water with tritium.

М	gO(light)	MgO(light)	MgO(light)
time (h)	1	4	24
concentration (Bq)	522066	522440	541768
removal ratio (%)	5.12	5.05	1.54
N	/IgO (heavy) MgO (hea	avy) initial
time (h)	4	24	
concentration (Bq)	533224	539834	550232
removal ratio (%)	3 09	1 89	

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CO5-5 Application of Neutron Activation Analysis to Micro Gram Scale of Solid Samples

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INTRODUCTION: Instrumental neutron activation analysis (INAA) is often used in cosmochemistry, where meteorites are the objects for INAA. Chondritic meteorites (chondrites) and iron meteorites contain relatively high contents of Co and Ir compared with those in the earth crust. As Co and Ir have high sensitivity in INAA, they can be good markers for the identification of such extraterrestrial materials. In NAA of chondrites, a few tens mg of specimen is commonly used. For such a case, a few hundred $\mu g kg^{-1}$ of Ir and a few hundred mg kg⁻¹ of Co can be reliably determined. When an extremely small size (e.g., micro gram) of samples such as micrometeor-ites recovered on the Earth surface and tiny particles re-turned from extraterrestrial asteroids are to be analyzed by INAA, the conventional INAA procedure used for a few tens mg is not suitable. For such tiny samples, neu-tron irradiation with high neutron flux and long irradia-tion time (namely, high neutron dose) is required. Re-cently we have presented the INAA procedure for micro gram scale of solid samples [1]. In this report, the degree of increase in isotopic abundance induced by neutron irradiation is described in detail.

RESULTS: As INAA is a nondestructive method of elemental analysis, the same sample once subjected to INAA can be reused for different analytical purposes. For such a case, effects caused by neutron irradiation are of concern. Major concerns are the induced radioactivity and the increase in isotopic abundance including the production of long-lived radioactive nuclides. Scientifically, the latter case is important by far and, therefore, is considered here. In order to make the evaluation of degree of the increase in isotopic abundance meaningful, used are the data from the experiment [2], where 1.66 mg of a tiny grain from the asteroid Itokawa was irradiated by neutrons under the condition (19 h-irradiation, ϕ_{th} : 8.2 × 10¹³ n cm⁻² s⁻¹, $f\phi$: 3.9×10^{13} n cm⁻² s⁻¹, and 5MW-operation). Assuming the elemental composition of bulk LL chondrite [3] for this grain, the number of produced nuclides with neutron irradiation was calculated based on the Monte-Calro simulation code (MVP 2.0) [4] and the reaction cross section data (JENDL-4.0) [5]. Both stable and unstable (radioactive) nuclides could be produced and the result is summarized in Table 1. In calculation, the position of control rod, temperature of the reactor core, and the combustion rate of nuclear fuel were all taken into consideration, because the neutron irradiation of the experiment [2] was performed near the reactor core and, therefore, these factors

affect the neutron energy spectrum. The produced nuclides in Table 1 are grouped into stable nuclides (²¹Ne, ²²Ne and ³⁸Ar) and long-lived radioactive nuclides (³⁶Cl, ²⁶Al and ¹⁰Be). These nuclides are typical cosmogenic nuclides and radionuclides produced by nuclear reactions triggered by cosmic rays and commonly detected in extraterrestrial materials like meteorites.

In Table 1, the calculated values are compared with numbers of corresponding nuclides observed in extraterrestrial samples. For stable nuclides (of noble gases), measured values for a different Itokawa grain are given for comparison [6]. It is well acknowledged that noble gases are extremely sensitive in mass spectrometry. As seen in Table 1, calculated values and measured values in the Itokawa grain are mostly comparable for all three nuclides. For long-lived radionuclides, concentrations in the Gold Basin L4 chondrite (a shower sample, UA-1188) [7] are given for comparison in Table 1. These data were obtained by accelerator mass spectrometry. The measured values are smaller or much smaller than the calculated values, with the difference varying by an order to six orders of magnitude. Evidently the reuse of neutron-irradiated samples should not be allowed for noble gas mass spectrometry and accelerator mass spectrometry for the study on noble gas nuclides and long-lived radioactive nuclides, respectively.

Table 1 Production of some cosmogenic nuclides from 1.66 μg of LL chondrite by neutron-induced reaction in INAA^a

Target nuclides	Reaction	Produced nuclides	Number of Produced nuclides	Number of nuclides (original)	
	Stable nuc	clides (noble	gas)-productior	1	
²⁴ Mg	(n,α)	²¹ Ne	$1.58 imes 10^8$	1.15×10^{7}	
²⁵ Mg	(n,α)	²² Ne	5.21×10^{7}	3.49×10^{8}	
³⁷ Cl	(n,γ), β ⁻	³⁸ Ar	7.97×10^{6}	1.78×10^7	
Radioactive nuclides-production					
³⁵ Cl	(n,γ)	³⁶ Cl	2.53×10^{9}	9.06×10^{2}	
³⁹ K	(n,α)	³⁶ Cl	3.24×10^{7}	9.06×10^{2}	
²⁷ Al	(n,2n)	²⁶ Al	1.30×10^{5}	3.73×10^{3}	
¹³ C	(n,α)	¹⁰ Be	3.03×10^4	2.07×10^{3}	
^a See text for detail					

"See text for detail.

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CO5-6 Chemical Variations of Miller Range 07710 (L4) Collected in the Ice at Miller Range, Antarctica

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INTRODUCTION: Most of meteorites are currently collected from cold and hot deserts. As these meteorites may have resided on the Earth's surface, they have experienced variable degree of terrestrial weathering. Terrestrial weathering obscures petrographic and chemical features, leading to misclassification and misunderstanding of geochemistry and cosmochemistry of these meteorites. In addition, misinterpretations probably result from sampling effect [e.g., 1] A large data set for chondrites were obtained by using 250-300 mg of sample [e.g., 2]. This amount of sample is not enough to obtain representative chemical compositions [1]. Therefore, we examine how terrestrial weathering and sampling problem affect chemical classification by using Miller Range (MIL) 07710 (L4) which was collected in the ice at Miller Range, Antarctica.

EXPERIMENTS: Eight different aliquots were taken from MIL 07710 and carefully ground in an agate mortar. Eight powder samples were analyzed by using INAA and IPAA at the Kyoto University Research Reactor Institute (KURRI). Neutron irradiations were successively perfomred two times (10 s and 4 hrs irradiations). For IPAA, we used a linear electron accelerator at KURRI. Electrons were accelerated by the linear accelerator to about 20 MeV.

RESULTS: Twenty seven elements could be determined by using both INAA and IPAA. MIL 07710 was grouped into L4 based on petrological and mineralogical studies [3]. The homogeneity of chemical compositions was found to be less than 10% (1 σ ; n=8) for most lithophile elements analyzed, In contrast, the heterogeneity exceeding 10% was found in siderophile (Co, Ni, Os, Ir and Au) and chalcophile elements (Se). This observed heterogeneity is considered to be due to sampling problem and/or terrestrail weathering. We investigated how this heterogeneity affect chemical classification of this meteorites. Fe/Mn and Fe/(Fe+Mg) ratios of eight different aliquots of this meteorite are plotted in Fig. 1. As seen in Fig. 1, two samples weighing about 1 g fall within the field of L, while Fe/Mn and Fe/(Fe+Mg) ratios for other samples with having less than 0.6 g fall in the ranges of L and LL. Eight samples display a positive correlation between Fe/Mn and Fe/(Fe+Mg) ratios. Such a trend can be explained by different modal abundances of metal and/or terrestrial weathering of metal. Jarosewich [4] compared major element abundances of fall meteorites with those of find meteorites from Antarctica and found that total Fe contents of Antarctic H chondrites are lower than those of fall H chondrites, but that total Fe contents of Antarctic L

and LL chondrites cannot be distinguished from those of fall L and LL chondrites. Therefore, terrestrial weathering could not explain such a positive correlation seen in Fig. 1. Fe/Mn and Fe/(Fe+Mg) ratios for mixtures of metal and olivine, and metal and low-Ca pyroxene were calculated, and the calculated results are shown in Fig.1. Different modal abundances of metal can explaine observed positive correlation among eight samples. It is concluded that metal is heterogenous distributed in sample weight less than 0.6 g.



Figure 1. Fe/Mn vs. Fe/(Fe+Mg) diagram for ordinary chondrites. Solid and dash lines represent mixing between metal [7] and low Ca-pyroxene [8], and metal [7] and olivine [8]. The proportional contributions of metal are shown as percentages among the mixing lines. Parentheses in sample position represent homogenized mass.

Siderophile element abundances are also useful for classification of ordinary chondrites [2]. Siderophile element abundances for most MIL 07710 samples analyzed in this study do not fall within the any range of ordinary chondrites. It is well known that sulfide and metal are more susceptible to terrestrial weathering than silicate [5] and that some Antarctic chondrites show fractionated abundance patterns for chalcophile and siderophile elements [6]. Therefore, observed scattering of siderophile element are attributed to terrestrial weathering.

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INTRODUCTION:

After the Fukushima Nuclear Power Plant (FNPP) accident in March of 2011, radioactive cesium 137 (¹³⁷Cs) was deposited over a large area in eastern Japan. Most of the ¹³⁷Cs remained in environmental soils and sediments, because of its 30-year physical half-life and strong adsorptive properties. A portion of the ¹³⁷Cs within these soils and sediments was transported downstream with suspended solids (SS). Additional sites affected by ¹³⁷Cs appeared downstream of the contaminated watershed (Sato *et al.*, 2013), and most ¹³⁷Cs had been accumulated in Ogaki dam, along with suspended matter which originated from the watershed soils (Kitamura et al., 2014). The purpose of this research was to quantitatively evaluate the form and distribution of ¹³⁷Cs in watershed soils and river/lake waters. Based on the measurement results, the factors facilitating the determination of the solid-phase ¹³⁷Cs concentration were discussed.

SURVEY and EXPERIMENT METHODS:

We collected samples from the Natsui River Basin (750 km²), located 40 km away from FNPP. The Natsui River is the water resource for the 160,000 people living in Iwaki city. River and lake waters, surface soils, and sediments were sampled using buckets, stainless shovels, and Ekman-berge sediment samplers, respectively. Surface soils from a depth range of 0-5 cm were mixed and combined from five point samples into a single sample.

The collected soil samples were desiccated by a freeze dryer(FD-81), and were then divided by particle size using sieves (2000, 850, 425, 250, 106, 75 and 25 μ m) after removing particles over 2 mm in diameter. Measurements of ¹³⁷Cs and organic carbon content (NC-22F) were carried out for the separated particles. Water samples (20 L) were separated into SS residue and filtrate through filtration, using a 1 μ m glass fiber filter (GF/B). The ¹³⁷Cs contained in the filtrate was concentrated into a disk (Empore TM Cesium Rad Disk) by forcible filtration. Afterwards, the ¹³⁷Cs in SS residues within the filters and in the disks were measured by a Ge semiconductor detector (GC4020/3020, CANBERRA).

RESULTS and DISCUSSIONS:

It was previously reported that ¹³⁷Cs in soils were dependent on the specific surface area of the soil particles (He and Walling, 1995). In order to analyze the adsorption characteristics of ¹³⁷Cs in soils, the ¹³⁷Cs (logarithm value) in each particle size category was plotted against representative particle size (logarithm value). A regression line slope of S=0 indicates that the ¹³⁷Cs was independent of particle size, while S =-1 indicates that the soil ¹³⁷Cs was dependent on the specific surface area of all

particles. Regression line slopes from the forest soil samples ranged from -0.8 to -0.4. This suggests that the ¹³⁷Cs in forest soils tend to be relatively dependent on the specific surface area. Conversely, regression line slopes from the bottom sediment samples in dam and paddy soils were almost zero. It was considered that the ¹³⁷Cs was equalized between different particle sizes, because the soil particles were more frequently exposed to water.

 137 Cs in each particle size, plotted against the organic carbon content in the forest soil at upstream site (Fig. 2). The result showed there was a strong correlation between these variables, and that the 137 Cs per organic carbon (*i.e.*, regression slope) was relatively constant (2,500 Bq/g -organic carbon) regardless of the season. This value was strongly related to the 137 Cs deposition, indicating that the organic carbon was an essential qualitative factor influencing 137 Cs.

Ratios of particulate ¹³⁷Cs in water were about 90% in upstream site, and about 20% in Kodama dam site, even during the normal river water stage. These results suggest that SS, as a carrier substrate, had a significant impact on the total ¹³⁷Cs loading to the sea. Calculated K_d values (¹³⁷Cs in SS residue divided by the ¹³⁷Cs in filtrate) were distributed from 0.066 to 62 ML/kg. Because there was not enough time to dissolve from particulate ¹³⁷Cs to dissolved one, the K_d was significantly higher than those values of previous studies. These results indicate that the behavior of ¹³⁷Cs in the natural environment is dominated by the movement of particles during floods, as well their adsorption on organic matter particles.







Fig.2 Relationship between ¹³⁷Cs and the organic carbon **REFERENCES:**

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Study of Localization Estimation of Abasic Sites in DNA Irradiated with Ionizing Radiation

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INTRODUCTION:

DNA lesions induced by ionizing radiation and chemicals can cause mutation and carcinogenesis. In particular, "clustered damage" site, that is a DNA region with multiple lesions within one or two helical turns, is believed to hardly be repaired. This damage is considered to be induced, e.g., around high-LET ionizing radiation tracks. However, detail of the damage is not known. We have already developed a method for estimating degree of localization of abasic sites (APs) in DNA using Förster resonance energy transfer occurred between different fluorescence probes ("hetero-FRET" using Alexa350 and Alexa488) [1]. The results showed that ${}^{12}C^{5+}$ beam produced close APs within a track: the apparent distance calculated was approximately 17 base pairs [2]. This finding indicates that *direct radiation effect* of ¹²C⁵⁺ beam near the Bragg peak produces clustered DNA damage. We have recently applied the method to DNA in a cell-mimetic radical scavenging condition [3,4]. However, there are some problems of the complex protocol and of the sensitivity due to the low extinction coefficient of Alexa350. We have, therefore, developed "homo-FRET" occurred between two or more Alexa488 molecules. We will obtain magnitude of FRET also from "fluorescence anisotropy" of homo-FRET between Alexa488 molecules. The new protocol using homo-FRET enables us to estimate DNA damage localization without any enzymes and improves sensitivity to detect a clustered damage.

EXPERIMENTS:

•Sample preparation and γ -irradiation

The plasmid DNA digested by Sma I was used (linear form). The DNA was dissolved to be 0.1 g/L in 0.2 M Tris-HCl buffer (pH 7.5) which is a cell-mimetic condition in relation to radical scavenging capacity. Twenty microliters of the DNA solution was transferred to a microtube (0.5-mL size), and was irradiated with ⁶⁰Co γ -rays (LET: ~0.2 keV/µm; Kyoto University Research Reactor Institute: KURRI) as a standard radiation source.

•*Preparation of fluorophore-labeled irradiated DNA and the FRET observation*

The irradiated DNA (10 μ L in water) and 10 μ L of 100 mM Tris-HCl (pH 7.5) were mixed in a microtube. Two microliters of Alexa488/DMSO was added to the DNA solution and was incubated for 24 h at 35°C. The fluorophore-labeled DNA was purified by ethanol-precipitation followed by ultrafiltration. The fluorescence anisotropy was measured at 525 nm (ex. 470 nm).

The anisotropy, $\langle r \rangle$, is defined as follows:

$$< r > = rac{I_{VV} - G \bullet I_{VH}}{I_{VV} + 2G \bullet I_{VH}},$$

where I_{VV} is the fluorescence intensity when the excitation and emission polarizers are both vertically oriented. I_{VH} is one when the excitation/emission polarizers are vertically/horizontally oriented. *G* is the grating factor defined as I_{HV}/I_{HH} .

RESULTS AND DISCUSSION:



Fig. 1. Relationship between AP average density (the number of APs per base pair) and fluorescence anisotropy for ⁶⁰Co γ -rays (•). The solid line indicates a theoretical curve for Poisson distributed APs in DNA.

In general, fluorescence anisotropy decreases with increasing FRET [5]. As shown in Fig.1, there was a little difference between the γ -ray data points and a theoretical curve based on Poisson distribution. This finding suggest that APs produced by the γ -rays are likely to be localized compared to those randomly distributed. This tendency is similar to the hetero-FRET results as shown previously [2-4]. A radiation "spur" on or nearby DNA might sometimes produces clustered damage. In any case, knowledge of AP distribution for ⁶⁰Co γ -rays *as a reference* is quite important to study DNA damage by ionizing radiation.

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CO6-2 Structural Insights of the Degradation Scheme of Yeast 20S Proteasome Studied by Genetic Mutation and AFM Motion Imaging

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INTRODUCTION: Proteasomes are the multicatalytic protein complexes with large molecular weight. It is well known that the ubiquitin proteasome system plays an important role in regulated proteolysis. Proteasome works as 26S particle that is composed of two 19S regulating components and a 20S proteasome. The 20S proteasome forms barrel shape and is composed of four rings, α - and β -rings; each ring contains highly homological seven α and seven β -subunits. Each of 1, 2 and 5 β -subunit has different enzyme activity; 1 is caspase-, 2 tryptic- and 5 chymotryptic- like activity, respectively. It was found recently that inhibition of 20S proteasome activity resulted in decrease and disappearance of cancer cells. The complex structure of 20S proteasome and its inhibitor have been determined by X-ray diffraction method. Binding site of inhibitor is closed to Tyr170 and Thr1 of the β 5 subunit. It is considered that an aromatic ring of the inhibitor interacts with Y170 by ring-stacking force. Since weak interaction of compounds to active amino acid residues we found, processing or degradation assay have been clarified by use of genetic mutations of the yeast 20S proteasome. Tyrosine mutations to Ala, Phe and Leu may not have interaction with the inhibitor and strictly changes binding activities to the 19S particle. The whole particle, 26S proteasome, including such 20S core is very important in the actual living cell for a degradation or stabilization on the cell circulation. Especially, a motion or behavior of the 26S proteasome when a regulatory particles bound should be clarified at atomic- or nano-scale resolution, we have tried to visualize such behavior as a motion capture of the atomic force microscopy (AFM) technique.

EXPERIMENTS: Yeast 20S (wild, and genetic modifications in the β 5 subunit; Y170A, Y170L and Y170F) and 26S proteasome particles were prepared by using yeast (*Saccharomyces cerevisiae*) cell cultivated. The crude extract was purified by using M2 affinity and Mono-Q anion exchange chromatography techniques. When the 26S particle was isolated, gel filtration chromatography was continuously applied with a buffer solution including ATP. Isolated 20S proteasome was concentrated by ultrafiltration. Activity measurement of wild and mutant proteasomes in the presence of the inhibition was carried out in order to reinvestigate the inhibition

mechanism. AFM images of the 26S proteasome were measured by NanoExplorer at 20°C at the Research Institute of Biomolecule Metrology in Tsukuba. Particle analysis was carried out by AFM reconstitution.

RESULTS: Genetic mutation for Y170 would have activities because of a lack of aromatic ring in the amino acid residue in the Ala and Leu, binding ability of the inhibitor might be lost. Typical activity assay for wild-type 20S shows reduction, otherwise the Y170L has no decrease of activity assay. Since additive order or combination of substrates and inhibitors affects hydrolysis strongly, 26S whole particle may have specific character in terms of own-shape.

The AFM measurement shows whole structure of the 26S particle, and analyses of resultant images describe a shape and size, and also a structural motion. Intensity distribution analysis, which reconstitutes outer shape of particle by enormous amount of omnidirectional image data, was applied for the AFM image that is able to judge as 26S proteasome. Intensity distribution of AFM data (Figure 1a) shows symmetrical shape, which is consistent with the size and shape of a double-capped whole 26S particle composed of central 20S and the both ends (Figure 1b). Measurements of AFM images of proteasome in the presence of substrates are in progress in order to analyze dynamic motion when proteasome is working.



Figure 1. Distribution analysis of AFM data of 26S proteasome.

a) Intensity distribution of 26S particle of AFM image.

b) Molecular model of 26S proteasome particle.

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Introduction

Boron neutron capture therapy (BNCT) is an attractive therapy for local tumor control in the treatment of brain tumor, melanoma, and so on [1,2]. However, some important issues are remained: the tumor-specific accumulation of highly concentrated boron, the real-time quantification of boron concentration in local tumor tissues, and the prevention of re-growth and metastasis of residual tumor cells after BNCT. Recently, much progress has been seen in tumor immunotherapy, and attracts many attentions of clinical researchers [3,4]. To achieve successful tumor regression, we try to establish the combination treatments with BNCT and immunotherapy. Using the radiotherapy with X ray irradiation, we firstly examined the anti-tumor effect of the combination therapy.

Results and Discussions

The establishments of cancer immunotherapy

In last year, we established some immunotherapies, e.g. blockade of immunological checkpoints with anti PD-1mAb and/or anti CTLA-4 mAb, vaccination with dendritic cells (DCs), and adoptive cell transfer therapy (ACT), using mouse model. Until now, we investigated the mechanisms of potent anti-tumor immune response by ACT under lymphopenic condition [5,6]. Assessing anti-tumor effect by anti PD-1 mAb or anti CTLA4 mAb, we confirmed the anti-tumor effect by blockade of immunological check points (Fig.1). In addition, similar to clinical trials, we found that anti-tumor effect was not sufficient for the tumor regression in some mice, whereas few mice treated with mAb could reject tumors. Using this protocols, we assessed whether anti-tumor effect is enhanced by the combination with radiotherapy.

Anti-tumor effect by the combination with radiotherapy and immunotherapy.

To assess the effect of combination therapy with radiotherapy and immunotherapy, tumor bearing mice were treated by mAb, and were locally irradiated by X ray (12Gy). Expectedly, significant tumor regression was observed in mice treated with the combination therapy (Fig.2). Analyzing anti-tumor immune response, we found that the induction of tumor antigen specific CTLs was not augmented by combination therapy, whereas local X ray irradiation lead to the increase of CTL population. Notably, compared with prior-treatment, later administration of mAb could not lead to significant anti-tumor effect during combination therapy, suggesting that the appropriate timing of mAb treatments is important for the induction of strong anti-tumor effect during the combination therapy. Based on these observation, now, we are trying to establish the effective combination therapy with BNCT and immunotherapy.



Figure 2. Anti-tumor effect by the combination therapy with local radiotherapy and the blockads of immuno-logical check points. 1x 10⁶ LLCgp33 cells were transplanted on right leg. Until 5 to 11 days, mice were treated by the immunotherapy with anti CTLA-4 mAb (left graph) and anti PD-1 mAb (right graph). Some mice were locally irradiated by 12Gy X ray, and tumor growth was assessed by the tumor area. The *p* values in the graphs indicate the statistical significance of the difference between each single treatment and combination therapy. Figures in the right of growth curve show the number of tumor rejected mice per total mice in each treatment group.

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[6] Suzuki T. et al, Cancer Immunol. Immun. 65(3): 341-345, 2016 T. Chatake, S. Fujiwara¹, Y. Yanagisawa² and I. Tanaka³

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INTRODUCTION: Recent neutron source had achieved rapidly advancing development in a quarter century. Especially, the pulsed neutron source at J-PARC had enabled us various neutron experiment, which could never been carried out due to lower neutron beam from neutron reactor. In the present project, the following the forefront experiments were studied for neutron biology. (1) D/H contrast neutron protein crystallography (Fujiwara) (2) The combination of scattering experiments of physiologically active substances derived from Bacillus subtilis *natto* (Yanagisawa), and (3) Dynamic neutron polarization technique for neutron protein crystallography (Tanaka). Here, the results of (1) and (2) are reported.

(1) D/H contrast neutron protein crystallography

EXPERIMENTS: Bovine pancreatic ribonuclease A (Sigma) was used for this study. Crystallization was carried out in H₂O-solvent crystallization solution, in the similar manner as reported earlier [1]. The obtained H₂O-solvent crystal (h-crystal) was converted to D₂O-solvent crystal (d-crystal) by vapor diffusion method. Neutron diffraction data sets both of d- and h- crystals were collected using BIX-3 diffractometer at JRR-3M nuclear reactor. So far, the contrast between D and H atoms was calculated in reciprocal space [2]. On the other hand, we designed another calculation method (real space D/H contrast method), and developed software for real space D/H contrast method. This method was applied for the two neutron data sets of the d- and h- crystals at 1.8 Angstrom [3].

RESULTS: The real space D/H contrast maps provided detailed pictures of the H/D exchangeable atoms in ribonuclease A and its hydration with two superior properties to the previous D/H contrast method. (i) all of the observed amplitudes can be used to visualize the D/H contrast, while only the amplitude commonly observed in both d- and h- crystals can be used in the previous method. (ii) real space D/H contrast map can be easily incorporated into standard neutron crystallography. These advantages contributed to interpretations of the 3D structure of this protein including hydrogen atoms with high accuracy.

Solvent region was also investigated using D/H contrast map. Fig. 1 shows average density in the D/H contrast map along the distance from protein surface, where the surface is accessible surface area. The results strongly suggested the 1st hydration shell present 1.4-1.6 Angstrom apart from the protein surface, and the density shape likes a donut with the width of ~1.2 Angstrom. The





Fig. 1. Density of solvent region in D/H contrast map.

(2) Combination of scattering experiments of physiologically active substance derived from Bacillus subtilis *natto*

EXPERIMENTS: Bacillus subtilis *natto* secrets abundantly water soluble vitamin K_2 (MK-7). The chemical and structural compositions of MK-7 have not been determined, while MK-7 has been sold as supplements. Our aim of this project is to elucidate structural composition by the combination of scattering method (DLS, XANS, and SANS). The scattering method requires MK-7 of high quality and quantity. In this physical year, procedure of purification was improved to achieve purification of MK-7 with the highest quality ever.

RESULTS: 20 mM Tris-HCl (pH7.6) buffer was used through purification. 1 L of Cultured medium was filtered and concentrated to 17.2 mL volume containing 0.3 M NaCl before purification as described before [4]. 1st purification was carried out DEAE FF (5mL) (direct injection) with gradient of 0.3 M -> 1.0 M NaCl. The main fraction was further purified using the 2 times of DEAE FF (1mL) with the same gradient. After ion-exchange chromatography, gel-filtration chromatography was carried out using sephacryl S-200 (ϕ 16x60cm). At the first gel-filtration, two peaks were observed in the elution curve. The former peak was collected and, they were purified in the same gel-filtration again.

Finally, 891 uL solution of purified MK-7 could be obtained. UV_{280nm} was 0.500. Bradford assay, in which BSA was used as standard, indicated 0.087 mg/mL protein was contained in MK-7. DLS and XANS are scheduled now. **REFERENCES:**

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CO6-5 Design and Synthesis of Boron-containing Compounds for Effective Accumulaion in Tumor Tissues and Detection by Boron Magnetic Resonance Imaging

S. Aoki, Y. Nishiura, T. Tanaka, Y. Hisamatsu, Y. Sawamoto, and 2, induced by Cu(II) in aqueous solutions. R. Araki¹, T. Saido², T. Suzuki³, K. Horie³, R. Abe³, S. Masunaga⁴, K. Natsuko⁴, Y. Sakurai⁴ and K. Ono⁴

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INTRODUCTION: (BNCT) is one of powerful therapies for local tumor con-MRI. Interestingly, the probe 2 undergoes faster detrol in the treatment of brain tumor, melanoma, and so on composition than that of 1 at 37 °C and neutral pH, [1]. Some critical issues are remained for the successful which was successfully detected on ¹¹B NMR and MRI BNCT, such as the tumor-specific accumulation of highly (Fig. 2) [4,6]. concentrated boron and the real-time detection of boron concentrations in local tumor tissues. In this study, we have designed and synthesized new boron compounds appended with glucose moiety and metal chelators for BNCT and B NMR (nuclear magnetic resonance) or MRI (magnetic resonance imaging) [2].

As for the boron compounds for more efficient accumulation in tumor tissues, we have synthesized several glucose-based boron compounds and assessed their tumor/blood (T/B) ratios [3].

As for B-MRI, some icosahedral carboranes derivartives, 1 and 2, which are composed of 10 boron atoms and 2 carbon atoms and has considerable thermal and chemical stability, were synthesized (Fig. 1). We discovered the first example of full deboranation reaction of o-carborane catalyzed by d-block metal ions under Fig 2. ¹¹B MRI and ¹¹B{¹H} NMR (128 MHz) spectra of physiological conditions [4]. Among various d-block 2 (2 mM) in DMSO/0.5 M HEPES (pH 7)/D₂O after inmetals tested, Cu2+ accelerates most efficiently to produce 10 B(OH)₃ from one molecule of carborane 0.2 mM, 1.0, and 2.0 mM) of Cu(II). deriva-tives.



Fig 1. Deboronation reaction of carborane derivatives 1

EXPERIMENTS and RESULTS:

Tumor accumulation of boronyl glucose compounds. Low toxicity of the synthesized compounds were confirmed by animal tests. Although the distribution of these agents in tumor cells are low, it was found that T/B ratios of some compounds are 2.3~2.4, which is better than that of BSH (0.5) [3].

¹¹B NMR and MRI change of carborane derivatives by decomposition reaction promoted by copper(II).

Decomposition reaction of 1 and 2, as well as our pre-Boron neutron capture therapy vious compounds [5] were followed by ¹¹B NMR and



cubation at different concentrations ((a)~(f) 0, 0.02, 0.1.

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CO6-6

Measurement of Transmittance Spectra of a Cryo-Sectioned Tissue of Brain Tumor C6 Model in the Sub-Terahertz Reagion-III

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INTRODUCTION: The accelerator-based radiation source in the millimeter and terahertz wave region has very attractive feature for the spectroscopy. Coherent transition radiation (CTR), which has been emitted from the short bunches of electrons at the KURRI-LINAC, has been used to observe the transmittance spectra of a sectioned tissue of raw brain tumor C6 model as a collaborate study in the research reactor institute, Kyoto university. The absorption spectra in the sub-terahertz region had been not so clear for the raw tumor tissue although Ashworth-PC. et al. [1] had reported for the excised human breast cancer by a terahertz pulsed spectroscopy observed at 320 GHz, which was estimated a longer relaxation time component of the induced electricity for water molecules [2-3] in the raw tumor tissue for three years at the linear analysis.

We also estimated what kind of water molecules become dominant in the viable and necrotic cancer regions by the different measurement method as an aim of 2D mapping study.

EXPERIMENTS: (1) Instrument of Near-field in tera-hertz region: The experiment was performed at the coherent radiation beamline [4] at the 40-MeV L-band linac of the Research Reactor Institute, Kyoto University. The width of the macro pulse and the repetition rate of the electron beam were 47 ns and 60 Hz, respectively. The charge of a bunch was 1.5 nC. The THz-wave source was CTR emitted from an aluminum foil with 15- μ m thickness. The radiation was detected by a liquid helium cooled Si bolometer. The conical cone with an aperture 260 μ m in diameter was used as the illumination probe and its F-number was 2.5. The spectrum of CTR was measured by a Martin-Puplett type interferometer. The schematic diagram of the experiment was shown in Fig.1.



Fig.1 The schematic diagram of the experiment.

(2) <u>Sample preparation</u>: A cryo-sectioned (thickness=100 μ m) tissue was prepared from the raw C6 glial tumor model using a Cryo-section Maker (Leica) and was sealed sandwich-type with saran-wrap film (thickness=10 μ m), under freezing condition (-20 C) before the measurements.

RESULTS: The photograph of the cryo-sectioned tissue with the measurement points was shown in Fig.2. The spectra were measured at every point. The 2-dimensional imaging of the observed intensity at the wavenumber of 19 cm⁻¹ was shown in Fig.3. The comparison between the spectral image and the H.&E.-stained Image is now progressing.



Fig.2 The photograph of the cryo-sectioned tissue with the measurement points.



Fig.3 The 2-dimensional spectral imaging at the wavenumber of 19 cm^{-1} .

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CO6-7 Protein Profiling and Drug Delivery on Cellular Apoptosis in Human Vitreous

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INTRODUCTION: Protein profiling in human vitreous may change in various vitreoretinal diseases. Regmatogenous retinal detachment is a severe retinal disease due to massive loss of retinal photoreceptors. Photoreceptor loss is mainly caused via molecular cascade to apoptosis. During this process, many proteins are released or accumulated in vitreous. Since photoreceptor apoptosis is initiated by activation of P2X7 receptor, P2X7 receptor blocker, brilliant blue G (BBG) may have therapeutic effects on retinal detachment. In this study, we examined protein profiling and drug delivery in human vitreous samples obtained during vitreous surgeries.

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EXPERIMENTS:

Human vitreous samples are examined by HPLC and the results are compared between controls and various diseases. BBG concentration was analyzed with LC.

RESULTS:

The analysis showed major reported vitreous protein in human samples. The time-dependent changes of BBG are also observed by LC. The BBG concentration in eye and blood was also compared in the dose-dependent manner. Further studies are needed for detailed analysis in vitreoretinal diseases.

CO7-1 A Fundamental Experiment for the Measure Against the Activation of the Irradiation-room Concrete at BNCT facility (II)

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INTRODUCTION: At present time, the research and development into several types of accelerator-based neutron sources for boron neutron capture therapy (BNCT) are underway by several research groups in the world, with Cyclotron-based BNCT Epi-thermal Neutron Source (C-BENS) at the head of the list [1]. In near future, BNCT using the accelerator-based neutron sources may be carried out at several places in the world. Unlike the facilities for radiation therapy and charged-particle therapy, the neutron yield is larger at BNCT facility. Then, the activation of concrete, which is a main structure of the irradiation room, is larger. The use of low-activation concrete is prefer in the viewpoints of the decrease of exposure under the work in the irradiation room, the decommissioning of the irradiation room, etc.. This research is intended to perform the characteristic estimation for low-activation concrete and confirm its usability at BNCT facility. In 2015, Kyoto University Reactor (KUR) was not operated. Then, a characteristic estimation was performed for the measure against the activation due to a neutron shield for concrete using an Am-Be neutron source, as the same manner in 2014.

METHODS: A characteristic estimation was performed for two kinds of resins such as Resin A and Resin B, and also Resin A containing B₄C, which are under development. As nine resin sheets of 10-cm side, 10-cm long and 2-mm thickness were stacked on the concrete surface, the shielding performance against the Am-Be neutron source was estimated by foil activation method. Indium foil was used as an activation foil. The shielding characteristic for fast neutrons was estimated by the activity change for In-113(n,n')In-113m reaction, and the generating characteristic for the secondary thermal neutrons was estimated by the activity change for In-115(n, γ)In-116m reaction.

RESULTS: Figure 1 shows the radioactivity changes dependent on the neutron-shield thickness for the In-113(n,n')In-113m reaction. Figure 2 shows the radioactivity changes dependent on the thickness for the In-115(n, γ)In-116m reaction. In these figures, the radioactivity changes for Resin A, Resin B and Resin A with B₄C are drawn. From the comparison for the radioactivity changes for the In-113(n,n')In-113m reaction, it was found that the shielding effect for fast neutrons was a little larger for Resin B than for Resin A. Also, it was found that the shielding effect for fast neutrons was a little smaller for the resin with B₄C, as its hydrogen density was smaller. From the comparison for the radioactivity the shielding effect for fast neutrons was a little smaller for the resin with B₄C, as its hydrogen density was smaller. From the comparison for the radioactivity the shielding effect for fast neutrons was a little smaller for the resin with B₄C, as its hydrogen density was smaller. From the comparison for the radioactivity changes for the In-115(n, γ)In-116m reaction, it was

CONCLUSION: When the KUR operation is restarted, the estimations for the important characteristics of low-activation concrete are planned, such as the shielding effect for neutrons and gamma rays, the generation of the secondary gamma rays, etc., using Heavy Water Neutron Irradiation Facility [2]. Also, the estimations for the characteristics are planned for short-life activation and long-life activation using Pneumatic Tubes.



Fig. 1. Radioactivity changes dependent on the thickness for the In-113(n,n')In-113m reaction.



Fig. 2. Radioactivity changes dependent on the thickness for the In-115(n,γ)In-116m reaction.

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Antitumor Effectivity by Gd-neutron Capture Therapy Using Gd-DTPA-incorporated Calcium Phosphate Nanoparticles

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INTRODUCTION: Gadolinium-157 has been getting attention as alternative for neutron capture therapy (NCT) agent because of its high thermal neutron cross section (255 000 barns), which the highest among all stable elements. Gadolinium neutron capture reaction (Gd-NCR) results in release of gamma rays, which reduce the localization effect of the treatment, which on the other side is increasing the possible additional effect if Gd-157 is accumulated to a bulk tumor cluster [1].

In this work, we performed histo-pathological examinations to evaluate the effects of Gd-NCT according to tumor growth suppression on multiple-injections Gd-DTPA/CaP nanoparticles in vivo.

EXPERIMENTS: *In vivo* evaluation was performed on colon-26 tumor-bearing mice irradiated for 60 minutes at nuclear reactor facility of Kyoto University Research Reactor Institute with average neutron fluence of 1.8×10^{12} n/cm²[2,3]. Antitumor effect was evaluated on the basis of the change in histopathological examinations using HE staining & TUNEL staining.

RESULTS: Tumor growth was suppressed until around four times of the non-treated group. Possibility of neutron depression for higher gadolinium concentration, there might be not sufficient amount of neutron reached deeper tumor site. Higher accumulation of gadolinium on tumor surface might reduce cancer cells killing effect at the tumor core[2,3].

Multiple-injection irradiated of bare Gd-DTPA group shows slightly better tumor growth suppression compared to control irradiated group. This proves low toxicity of GdDTPA/CaP nanoparticles, because all mice survived for both single and multiple injection groups.

Tumor cells were destroyed after NCT and changed into granulation tissue. Number of cells killed after treatment were similar for single and multiple injection groups of Gd-DTPA/CaP nanoparticles. Non-treated group shows normal histology with clear cytoplasm and nucleus(Fig.1)[3].

Evaluation of possible apoptosis occurred on cancer cells by detecting the DNA fragmentation following GdNCT treatment. Negative control for both irradiated and non-irradiated groups was also prepared during the apoptotic assay. Number of cells undergoing stained by TUNEL, which correlates to the number of apoptosis, was higher on GdNCT treated group compared to the non-irradiated ones(Fig.2)[3].



Gd-DTPA/CaP multiple injections + Irradiation

Gd-DTPA/CaP multiple injections

Figure 1. Evaluation of Antitumor Effectivity (H&E Staining)



Gd-DTPA/CaP multiple

injections + Irradiation

Gd-DTPA/CaP multiple injections

Figure 2. Evaluation of Antitumor Effectivity (TUNEL Staining)

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CO8-1

Development of Non-Destructive Methods Adapted for Integrity Test of Next Generation Nuclear Fuels at KURRI-LINAC

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INTRODUCTION: Fast reactor system with transuranium (TRU) fuels containing minor-actinides (MAs) is one of attractive options for the incineration of high-level radioactive wastes. In order to develop the integrity evaluation methods of the fuels for safety operation of the fast reactor system, the N-DeMAIN (Development of Non-destructive Methods Adapted for Integrity Test of Next Generation Nuclear Fuels) project has been started from 2014. In the project, the identification and quantification of nuclide in the fuels will be conducted by the neutron resonance transmission analysis (NRTA). Moreover, the determination of temperature distribution in the fuels based on the Doppler-broadening and neutron imaging are planned. KURRI-LINAC is used for the project because it is the only pulsed neutron facility where nuclear materials can be utilized in Japan. To achieve these purposes, The KURRI-LINAC neutron source should be improved and modified for the high quality neutron beam regarding neutron flux, time resolution and spatial resolution. Based on numerical analyses, the neutron source system, especially moderator, reflector and collimator, was newly designed and it was installed at KUR-RI-LINAC as shown in Fig.1. Then, the characteristic of the neutron source system was investigated.



Fig.1 New neutron source installed in KURRI-LINAC

EXPERIMENTS: Neutron spectrum was obtained by the Time-of-flight (TOF) method at 12 m beam line at KURRI-LINAC where the capture gamma-rays from ¹⁰B and dummy samples were measured. The samples were set at the 135 degree with respect to the incident electron beam and 12.7 m distance from the target. Two BGO scintillators (2 in. diam. \times 2 in. length) were used for measuring capture gamma-rays and arranged at 80 mm from the samples. The effective area of the detectors was

covered by lead (50mm thick) for reducig background gamma-rays. The operating conditions of the accelerator were as follows: average beam current was 16.3 μ A, frequency was 50 Hz and pulse width was 100 ns. The polyethylene was used as a moderator and its size was 15 cm square and 5 cm thick which was designed to increase the epi-thermal neutron flux. The collimator in the beam line was arranged in tapered shape using several polyethylene tubes containing boron which have 5, 10 and 15 cm hole, respectively. The size of outlet side of the neutron beam is 5 cm in diameter.

RESULTS: The measured TOF spectrum using ¹⁰B and dummy samples is shown in Fig. 2. The horizontal axis is neutron energy and the vertical axis is neutron flux per unit of time, unit of area and unit of average beam current. The values over 10 eV are distorted by the gamma flash effect from the neutron source. It was found that the gamma shield to reduce the effect is important for the measurement of epi-thermal neutrons. From the results under 10 eV region, the neutron flux was 1.9×10^2 $[n/cm^{2}/s]$ per 1 µA in average beam current. Note that the value of the neutron flux is slightly overestimated because the detection efficiency for 662 keV gamma-rays from ¹³⁷Cs standard source was used instead of that for 448 keV from ¹⁰B capture gamma-rays. In the case of the maximum power of KURRI-LINAC which is 6 kW, it was found the maximum neutron flux can be obtained 3.8 $\times 10^4$ [n/cm²/s] at 12 m beam line with the present neutron source system.



ACKNOWLEDGEMENT: The authors would like to thank Ms. Harada of Nagoya University for her contribution of the numerical analysis and valuable discussion. Present study includes the result of "Development of Non-Destructive Methods Adapted for Integrity test of Next generation nuclear fuels" entrusted to the Kyoto University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).
Stability of Novel Cyclic Monoamide Extractants for U(VI) against γ-Ray Irradiation in HNO₃

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INTRODUCTION: Uranium is the predominantly major component of spent fuels, and extractants with high selectivity and capacity to U(VI) and radiation stability in nitric acid media are desired in the world. Monoamide extractants have been studied for the purpose due to very little generation of the secondary waste resulting from the Above all, "branched monoamodes" spent extractants. have been studied almost exclusively so far, probably because of relatively easy synthesis[1]. On the other hand, extensive studies on pyrrolidone and its related derivatives, which are categorized into "ring monoam-ides", convinced us that some ring monoamides with high symmetry would exceed branched monoamodes on the above-mentioned properties[2,3]. Based on the back-ground, we focused on 1,3-dialkyl-2-pyrrolidone (DRP) (Fig. 1). In this study, fundamental stability of two DRPs, 1,3-dibuthyl-2pyrrolidone (DBP) and 1,3- dihex-yl-2-pyrrolidone (DHP), against γ -ray irradiation was investigated.

R' R



Fig. 1. Chemical structure of DRP.

EXPERIMENTS: DBP and DHP were synthesized by modifying the method shown in the reference[4]. For the sample for γ -ray irradiation to DBP and DHP, solutions consisting of 30 vol% DBP or DHP in *n*-dodecane as the organic phase and 3 or 6 mol/dm³ (= M) HNO₃ as the aqueous phase were prepared in a Pyrex tube. Irradiation was carried out by the ⁶⁰Co source up to 1.14 MGy at room temperature under ambient atmosphere, similarly to the earlier study[5]. Irradiated samples were analyzed by ¹H NMR. CDCl₃ containing a known weight of TMS which is a standard material was used as the NMR solvent for the organic phase. The residual ratio of MEC was calculated by the area ratio of each signal with that of TMS.

RESULTS: No distinguished appearance changes were observed for the irradiated samples. In a ¹H NMR spectrum, neat DRP shows several signals. The calculated residual ratios for DHP irradiated in 3 and 6 M HNO₃ are shown in Fig. 2, respectively, together with the relationship between the position of hydrogen in the structure of DHP and that of the signal. For the sample irradiated in

3 M HNO₃ (Fig. 2(a)), the residual ratios calculated through (A) to (C) in DHP show an identical trend, where nearly no decomposition occurred after irradiation at ca. 0.8 MGy. This suggests that the stability of DHP in 3 M HNO₃ is better than that of water-soluble *N*-butyl- pyrrolidone (NBP) with the residual ratio of ca. 85 % at 0.8 MGy, although the solution conditions of both compounds during irradiation are different in a precise sense with each other[3]. On the other hand, for the sample irradiated in 6 M HNO₃ (Fig. 2(b)), the calculated residual ratios were found to differ depending on the position of hydrogen atom in DHP. While the residual ratio calculated from hydrogen of the side chain (A) remains nearly 100 % after irradiation at ca. 0.75 MGy, those calculated from hydrogen in the ring ((B) and (C)) decrease down to 40 through 60 %. The residual ratios of ca. 50 % were in accordance with the results for NBP. These facts indicate that DHP possesses satisfying radiation stability at around 3 M HNO₃. Further detailed investigations are ongoing.



Fig.2. Dependence of residual ratio of DHP on dose; (a) 3 M HNO₃, (b) 6 M HNO₃.

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CO9-2 Search for the Ultraviolet Photons Emitted from ^{229m}Th in its Deexcitation Process

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INTRODUCTION: Thorium-229m lies in the first excited state of the ²²⁹Th nucleus and this excitation energy is extremely low to be 7.8 ± 0.5 eV [1] corresponding to vacuum ultraviolet (VUV) photon of which the wavelength is about 160 nm. Thorium-229m is expected to decay by the gamma transition and/or the internal conversion (IC) process. Because its excitation energy value is in the same energy scale with that of the chemical bond, ^{229m}Th would decay by IC process in some chemical forms and in the other forms, the IC may be hindered. Consequently, the decay process of ^{229m}Th is able to vary depending on its chemical form. This phenomenon is unique to ^{229m}Th and makes ^{229m}Th nucleus very fascinating. Recently, photon emissions from the ^{229m}Th sample implanted into MgF₂ glass were reported and its half-life was evaluated to be about 6 h [2]. However, it is controversial [3]. In our study, the results of the photon detection for the hydroxide and fluoride precipitates of ^{229m}Th grown for 2 h or 13 h were previously reported [4]: decaying photons were observed for 13-h growth samples. Here, the results of the photon detection for the ^{229m}Th precipitates after purified and the ^{229m}Th samples implanted into the MgF₂ glass are reported.

EXPERIMENTS: Thorium-229m precipitated samples were prepared by the method reported previously [4]. In this study the additional procedure to purify the ^{229m}Th was performed before ^{229m}Th was coprecipitated with Sm, assuming that the half-life of ^{229m}Th would be long: about several hours. After ^{229m}Th was separated from ²³³U, the ^{229m}Th eluent was heated to dryness. Then, 8 M HNO₃ solution was added and ^{229m}Th was adsorbed on the ani-on-exchange resin (Muromac 1×8 200-400 mesh, 1 mL). The daughter nuclides of ²²⁹Th were removed by pouring 8 M HNO₃ solution. After that, 2 M HCl solution was passed through the ^{229m}Th column and ^{229m}Th was eluted. In the case of the ^{229m}Th samples implanted into MgF₂

In the case of the ^{229m}Th samples implanted into MgF_2 glass, ^{229m}Th was collected as the recoil product from the electrodeposited sample of ²³³U in vacuum.

RESULTS: On the results of the photon detection for the ^{229m}Th samples after purified, there was no decaying photon emission. In addition, on the result of the long-time measurement, the count rate of photons increased as time passed. This result is shown in Fig. 1. These growing photons agreed with the growth of the daughter nucleus ²¹²Bi. Taking into account the result in ref. 4, we concluded that no significant photons from ^{229m}Th were observed although we used about 200 mg of ²³³U: this amount was about 400 times larger than that in the study in ref. 2.

Because in the experiment on the ^{229m}Th precipitates,

VUV photons deriving only from the daughter nuclides were observed, we measured the photons emitted from the same 229m Th samples as those in ref. 2 to search the photons derived from 229m Th in MgF₂. On the result of the MgF₂ glass samples, decaying photons probably originating from multiple components including the daughter nuclides were observed. This result is shown in Fig. 2. We are precisely analyzing these data to elucidate these origins.



Fig. 1. The photon count for the ^{229m}Th precipitate (hydroxide form) grown for about 13 h. Black dots denote the photon count rate of the ^{229m}Th sample, solid curve describes the growth of ²¹²Bi and dotted line describes the count rate of background.



Fig. 2. The photon count for the 229m Th implanted into the MgF₂ glass for about 26 h. Black dots denote the photon count rate of the 229m Th sample and the solid curve describes the fitted decay curve consisted of two decaying components: these half-lives are about 10 h and 7 d.

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CO12-1 Basic Research of an Intense Terahertz Light Source Based on Coherent Cherenkov Radiation Matched to the Circular Plane

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INTRODUCTION: One of the author proposed an intense terahertz (THz) light source based on paralleled coherent Cherenkov radiation (CCR) which was generated by a relativistic electron beam traveling through a hollow conical dielectric [1]. As superimposing coherent diffraction radiation with many diffraction elements, the intense THz light can be generated by matching the CCR to the circular plane. We report results of the preliminary experiments of the CCR matched the circular plane (CCR-MCP).

PRINCIPLE OF CCR-MCP: The CCR intensity increases proportionally to the length of the electron trajectory in the dielectric [2]. The CCR power obtained with a long dielectric tube can be higher than power of coherent transition radiation (CTR). However, the CCR is emitted onto a conical surface whose apex locates at the electron beam, so that it is difficult to form a plane wave of the CCR beam with matching the phase. Then, a hollow conical dielectric with its apex facing the incident electron beam was proposed to overcome this difficulty [1]. By setting an angle between the generatrix and rotation axis to be half of the Cherenkov angle, all CCR that is reflected by the conical surface is in phase, forming CCR-MCP on the base of the hollow conical dielectric. When the absorption coefficient and the refractive index of the dielectric are expressed as α and n, respectively, the CCR-MCP beam saturates at a certain length L_c , which is given by

$$L_{C} = \frac{n\beta}{\alpha} \ln \frac{1+n\beta}{n\beta},$$

where β is the ratio of the electron speed to the speed of light. Because it is easy to focus a plane wave, more powerful THz light can be obtained by using a hollow conical dielectric with very low absorption coefficient.

EXPERIMENTS: We performed preliminary experiments of the CCR-MCP with an L-band electron linac at the Kyoto University Research Reactor Institute (KUR-RI-LINAC). Figure 1 shows the schematic layout of the CCR-MCP experiments. We used high-density polyeth-ylene for the hollow conical dielectric. The length and inner diameter of the hollow conical dielectric were 83 and 10 mm, respectively. It was located at 0.4 m from the aluminum foil mirror upstream. An aluminum collimator, whose length and inner diameter were 150 and 8 mm,



Fig. 1 Schematic layout of the CCR-MCP experiment in the KURRI-LINAC.



Fig. 2 Spectra of the CCR with the CTR.

was installed in front of the hollow conical dielectric not to expose the hollow conical dielectric to the electron beam. The electron energy was 42 MeV, and the charge in a micropulse was approximately 40 pC.

RESULTS: The CCR-MCP beam was observed with CTR beam generated at the aluminum foil mirror by a Martin-Puplett type interferometer. When the hollow conical dielectric was turned in the opposite direction, only the CTR beam was observed. Figure 2 shows the measured spectra of the CCR-MCP and CTR beams. The CCR-MCP intensity was strong three times of the CTR intensity. Our experimental results suggest that one can produce an intense THz light source based on the CCR-MCP with using more compact and high-charge electron beams.

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Effective Measures on Safety, Security, Hygiene and Disaster Prevention in Laboratories

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INTRODUCTION:

Important research aspects can be found in the following keywords such as safety, security, hygiene and disaster prevention. Nuclear research reactor is one of representative facilities together with these keywords under their operation. It is effective to investigate the latest status on practical measures on these keywords in various facilities including nuclear research reactors, to compare each other among facilities, and to discuss more optimized ones for our positive safety management. Through this process, it is also essential to investigate the latest international and/or national regulations and the movement of revision of them. This total discussion contents and its fruit are directly useful for all relating laboratories.

RESEARCH APPROACH:

General research approach is as follows.

- Measures of safety management during operation or standstill status of the real facilities would be investigated. This information would be used for our research discussion on the positive and more optimized safety management.
- It would not be a single year research, but maybe two to three years research for one theme.
- Information source of facilities would not be only KUR, KUCA or the other facilities in Kyoto University, but also the Kinki university research nuclear reactor or the facility of National Institute of Fusion Science, etc. This research is an active joint-research with these relating facilities and positive researchers on safety management.
- One of the distinctive features of this research is to involve office staffs as cooperators as well as researchers and technical staffs. In The University of Tokyo, most of the members in Division for Environment, Health and Safety are office staffs who knows real situation of safety management in laboratories very well.

Discussion target in FY of 2015 was determined as "safety control in use of small amount nuclear materials" through our member discussion. When using small amount of uranium (U) or thorium (Th) up to 300g (U) and/or 900g (Th), most of safety control procedures requested as radioactive materials are exempted by related regulations. We investigated the real situation of usage

and safety control of small amount of U and Th in Japanese laboratories. In addition, training and education to safety managers, users and officers relating small amount of U and Th were also discussed.

LATEST SITUATION ON USE & SAFETY CON-TROL OF SMALL AMOUNT of U & Th:

The following information was obtained by means of questionnaires which were sent to 37 users of small amount of U and Th in Japan.

<u>Usage</u>

Among 33 users who answered the questionnaires, it was 24 users to really use U and/or Th. 9 users do not use and just storage the materials in their laboratories. 17 users answered that they used the materials for the dyeing of the bio-sample in the electron microscope observation. Safety Control

16 users among 24 handled the small amount of the materials in general areas without setting specified areas. Radiation safety control is not requested in the Japanese regulation for the materials, however about half of real users take some safety measures.

Waste

18 users among 24 keep and storage their nuclear wastes in their facilities. There were many opinions in hope of collection of the waste by the government to dispose them. Hesitation and confusion were seen in some users about the consumption and the storage disposal of the nuclear materials. Information sharing on the waste management is essential.

NEXT RESEARCH TARGETS;

We started to discuss on the education framework, curriculum and textbooks for safety managers and users as well as relating officers to fill up the nuclear material controlling sheet for the use of small amount of U and Th. This discussion and establishment should be continued next year.

We express our gratitude for their strong support and active discussion of Dr. K. Yasuda (Kyoto University), Dr. M. Takahashi, T. Saze (National Institute of Fusion Science) and Dr. H. Yamanishi (Kinki Univesity) and others.

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Test of a Microcell Multi-Wire Proportional Chamber for a Muon-Electron Conversion Search Experiment, DeeMe

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INTRODUCTION: Discovery of Higgs boson filled the last piece of the Standard Model (SM) of elementary particle physics. The next coming issue is the physics beyond the SM (BSM). A discovery of mu-e conversion may be a clear clue to the BSM. DeeMe is going to search for mu-e conversion in J-PARC. The detector is required to tolerate to prompt burst pulses with an instantaneous hit rate of approximately 100GHz/mm² and width of 200ns and to detect electron signal with delay time of O(μ s) from the burst pulse. We have invented a new technique of dynamic gain control of wire chamber to avoid a long dead time by space charge effect.

EXPERIMENTS: Experimental setup is shown schematically in Fig. 1. Electron beam collimated to 18mm x 20mm penetrates a Multi-Wire Proportional Chamber (MWPC) and beam counters. Beam rate is tuned changing current of electron gun heater.



Fig 1. Experimental setup

The pulsed electron beam with tuned beam rate generated from the beam gun emulates the prompt burst pulse. Delayed electron signals are emulated by field emission electrons.

DC high voltage (HV) is applied to anode wires. MOSFET based switching module makes a pulsed HV to be applied to potential wires of the MWPC. During a main pulse comes, HV is applied to potential wires to make voltage difference between wires 0V. Soon after the main pulse passes the MWPC, potential wires are switched to ground level to make large voltage difference between anode-potential wires to detect delayed electrons. Large current is induced on the cathode strip readouts by the rapidly changing voltage on the potential wires. Amplifier should cope with this input. Fig. 2 shows the circuit of our amplifier. We tuned the circuit elements not to make the amplifier saturated by the large current input. Pole zero cancelation circuit is implemented between 2-staged amplifiers to compensate slow tail by ion movement. We have manufactured the final version of the chamber and the amplifier. This experiment was planned to be the first test of the final detector system under the condition equivalent to the final DeeMe condition.



Fig 2. Circuit diagram of the amplifier

RESULTS: The chamber is operated with a gas mixture of Ar/C_2H_6 . We went in a bit too much of a hurry, and exchanging the gas was not enough before applying HV on the chamber. Discharge cut all the wires of the final chamber. This told us importance of removing air contamination.

We switched the purpose of the experiment to a test of amplifiers. We replaced the broken chamber to a prototype chamber and continued the test. Amplifier test was good. The large current input by HV switching and burst electron pulse did not saturate it. The baseline successfully recovered for the delayed electron.

But we encountered another problem of applying nominal HV on the prototype chamber. A balance of electrostatic force and restoring force by wire tension determines wire position. The restoring force is stronger, so the wires stay the nominal position. But the difference of the strength is calculated to be small. High intensity electron beam may distort the balance, and cause wire position displacement a few times per some tens of minutes. Discharge occurred when wires get too close.

This gave us a hint of operating the chamber stable. In this experiment, we made the voltage difference between anode-potential wires 0V just during the burst pulse. We take data with cycle of 25 Hz and the data-taking period is just a few 10 micro-s. Attractive force between anode-potential wires made the wire position unstable. Changing the scheme of applying HV can solve this problem. Making the voltage difference just during the data-taking period, repulsive force works most of the time despite of the data-taking period. This repulsive force keeps distance between wires. Attractive force during the data-taking period is just a few 10 micro-s and the impulse is small enough to keep the wire position stability. Based on this consideration, we developed a new scheme of switching HV after this experiment. This successfully worked to make the operation of the chamber with nominal HV stable.

CO12-4 In-situ Analysis of Chemical Reaction between Phosphoesters and Iron Surface by Attenuated Total Reflection Infrared Spectroscopy and Neutron Reflectometry

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INTRODUCTION: In recent years, reduction of friction and wear in machines is one of the most important subjects from the viewpoint of global environmental issues, energy conservation and resource saving. Especially development of materials having excellent tribological properties is required to extend the life of machine parts by reducing friction loss. Particularly, under the boundary lubrication regime, oiliness additives and extreme pressure agents are used to reduce friction and wear. Extreme pressure agent is commonly known to form a 'boundary lubrication film' onto the metal surface via the chemical reaction by the heat generated in the friction process. The film prevents direct contact between metals, reducing the possibility of occurrence of seizure. To investigate the formation process of boundary lubrication film formed by extreme pressure agent on the metal surface, attenuated total reflection infrared spectroscopy (ATR-IR) and neutron reflectometry (NR) were used in this study.

EXPERIMENTS: As the sample substrate for ATR-IR, a germanium hemispherical ATR crystal which coated thin film of iron on flat surface was used. Poly-α-olefin and phosphate ester were used as base oil and extreme pressure agents respectively. Eight kinds of phosphate ester were selected for the study; mono-oxyl phosphate ester(mono-C8(straight chain)), di-oxyl phosphate ester(di-C8(straight chain)), mono-2- ethylhexyl phosphate ester(mono-C8(branch)), di-2-ethylhexyl phosphate esphosphate ter(di-C8(branch)), mono-dodecyl ester(mono-C12(straight chain)), di-dodecyl phosphate ester(di-C12(straight chain)), mono-octadecacyl phosphate ester(mono-C18(straight chain)), di-octadecacyl phosphate ester(di-C18(straight chain)).

First, time dependence for the reaction between phosphate ester and iron was investigated measuring the change in peak made by the reaction between iron and phosphate ester. The IR absorbance profiles obtained for 20 hours per one hour. When the monoester of non-mono-C18 was used, the peak was increased with time, and the peak value was the largest in all samples (Fig. 1). NR also proved the existence of boundary film formed by the adsorption of monoester onto the iron surface (Fig. 2(a)). On the other hand, when the diester and mono-C18 were used, the peaks did not increase with time. NR also proved that the boundary film was not formed onto the surface (Fig. 2(b)). It means that all of phosphoric esters were not adsorbed onto metal surface and the adsorption characteristics depended on the molecular weight.

Second, time dependence for the reaction between phos-

phate ester and iron at high temperature was investigated to understand the heat effect. Heating temperature was set to be 50, 100, 150, 200, and 250°C and heating time was 30 minutes. When the monoester was used, the peaks from the phosphoric acids increased, and the peaks became larger than the peaks measured at room temperature. When the diester was used, the peaks from the phosphoric acids did not increase.

From these studies, the phosphate ester with short alkyl chains without branch easily adsorbed onto metal surface, and their reactivity increased by heat.



Fig. 1. IR absorbance spectrum from the interface between iron and lubricant containing mono-oxyl phosphate ester (mono-C8(straight chain))





CO12-5 Single Event Test on On-board Computer for Nano Satellite "Horyu-4" using ²⁵²Cf

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INTRODUCTION: Kyushu Institute of Technology (KIT) has been developing nano satellites as Horyu-series since 2010. The main mission of Horyu-series is the high voltage demonstration by solar array and the Electro Static Discharge (ESD) experiment on orbit. Horyu-2 launched in 2012 suffered an anomaly by Single Event Latchup (SEL) on the On-board Computer (OBC). As a result, Horyu-2 eventually could not respond to commands from the ground station[1]. For the next satellite, "Horyu-4", the project members recognized that a counter measure for SEL is necessary to avoid the same fate as Horyu-2. The project members decided to conduct radiation testing using ²⁵²Cf at Kyoto University Research Reactor Institute. Radiation testing was performed twice in 2015. We used the circuit boards of Horyu-4 including OBC, Electrical Power System (EPS) and Communication and checked the failure modes caused by SEL. EPS provides 3.3 V and 5.0 V lines to each sub-system and it controls the reset system. The purpose of this test is to improve the stability of Horyu-4 under the effects of radiation. This document reports the detail of testing and results.

EXPERIMENTS: Figure 1 shows the experimental set-up. Circuit boards used in this test were EM and FM models for Horyu-4. These boards have as microprocessors H8 and PIC. The plastic package of microprocessors was removed so that the core of the microprocessors was directly exposed to ²⁵²Cf source. The circuit boards were set in a vacuum chamber. The pressure during the test was approximately 30 Pa. The voltage and current fed from the power supply were measured by a DAQ and oscilloscope and the operation of the microprocessors was verified by a PC with RS232 communication. An over-current protection (OCP) was implemented in the power line between each sub-system and EPS in the circuit boards. Once OCP detects over current due to SEL on the OBC. OCP sends a reset signal to EPS. To recover from the hang-up state of OBC by SEL, EPS automatically cuts the current from EPS to OBC. In this experiment, ²⁵²Cf source was mounted on XYZ stage shown in Fig. 2 and its position and height were controlled by a stage controller. The distance from microprocessors to the radiation source was from 10 mm to 30 mm.

RESULTS: The test procedure is as follows.

- 1) Check operation of the circuit board
- 2) Move 252 Cf source to above the microprocessor.
- 3) Monitor the system by PC.

The first test was conducted in August 2015. In the first test, the OCP seems to be able to detect the over current induced by SEL on the OBC and the reset signal was sent

to EPS. However, our measurement system could not acquire the recovering OBC because the reset signal from the OCP was too fast. For the second test, we improved the measurement system. The second test was conducted in September 2015. We observed that EPS could not cut the current feeding to OBC even though OCP properly detected over current. As a result of the investigation, a leakage current from 5.0 V line to 3.3 V line affected the reset system when EPS cuts the current of 3.3V line to OBC. To execute a proper reset, EPS must cut the current of 5.0 V line, too. We improved the configuration of the reset system and confirmed the reset system.

The improved Horyu-4 system was verified by this radiation test. Horyu-4 was launched on Feb 17th 2016 and achieved its main mission as the world's first acquisition of ESD waveform and image on orbit[2].



Fig. 1 Test Setup

XYZ stage



Fig. 2²⁵²Cf source mounted on XYZ stage

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CO12-6 Ethnographic Study on Nuclear Reactor - Radiation Hazard and Risk Management for Evacuee's Future in *Fukushiama* -

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In order to know about nuclear reactor in layman term, it might be highly helpful to describe it as ethnography by investigating how it has been concerned in human civilization. In this report we describe ethnography of evacuees from Fukushima Nuclear Accident in order to recover and live together in low dose radiation field as a different culture, in other words to know about it's various aspects and complicated conditions in constructing their new culture.

Topics of this progress report: We investigated the complicated difference between physical dose of radiation exposure (hazard) and sensibility feeling (risk) through scientific field works in Fukushima. And we discuss about the possibility to live in evacuee's hometown contaminated with low dose radiation (mainly ¹³⁷Cs).

It is essentially very difficult to apply anthropological study in Fukushima from an ethical standpoint. Fortunately, we overcome this difficulty by applying scientific participation in refuges to keep good relationship with refugees. Figure 1 shows radiation dose of an evacuee who lives in refuge and regularly visit his home in contaminated area with low dose radiation. His average radiation dose is 1.97μ Sv/day in his refuge, but it rises to 6.70μ Sv/day during stay in his home.



Figure 1. Daily fluctuation of radiation dose determined by a personal dosimeter (miniDOSE, Ray Systems Co.) H: at home, R: at refuge.

He says, "It's more healthy to work in my contaminated yard than being in refuge for my safety". Fortunately he has regulated his radiation dose to be under regulated limit, <1mSv/year, by managing the period of home-stay and also the amount of edible wild plants cropped in his contaminated mountain. Figure 2 shows the radiation surface dose (1m high from the ground), the dose vary with altitude and configuration.



Figure 2. The radiation dose distribution in the mountain yard. HOME:aroung 0.3 μ Sv/hr, RELATIVE HOT area in the mountain yard: 0.9-1.3 μ Sv/hr. Date from Fukushima Project, measured by Hot Spot Finder, HSF-1, Nihon Shahei Giken, Co.

He says. "I will eat the delicacies of the seasons up in mountain because I will miss it behind the season." He can calculate a limited intake amount of delicacies to be under safety level. This case shows that possibility of living in contaminated hometown by analyzing the hazard and accepting the risk.

On the very beginning of Fukushima Nuclear Accident, why scientist's statements about radiation hazard & risk were so different among them? There is no doubt that we scientists are responsible to such situation. Our next concerns about *Fukushima* in this research are scientific participation and inspection on the actor network among nuclear scientists.

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CO12-7 The XEP-e (eXtremely high Energy Plasma/ particle sensor for electron) of the ERG Satellite

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INTRODUCTION: The radiation belt called the Van Allen radiation belt exsists within the geospace that is the region of outer space near the Earth. It often causes a satellite's trouble. The Exploration of energization and Radiation in Geospace satellite (ERG) that is the JAXA's project will be lanched from the Uchinoura Space Center in 2016FY. This project aims at elucidating how highly charge electrons have been born while they generate and vanish repeatedlly along with space storms caused by disturbance solar wind. This satellite has four instrument parts, a Plasma Particle Experiment Suite (PPE), a Magnetic Field Experiment (MGF), Plasma Wave Experiment (PWE) to measure radiation particles, magnetic and electronic waves. The eXtermely high Energy Plasma/particle sensor for electron (XEP-e) is one of the PPE. It aims at measuring electrons from 400keV to 20MeV. We are now developing its Flight Model and used the KURRI-LINAC (Kyoto University) to calibrate it over 2 MeV electrons.

Table. 1. Mission Profile of the ERG

ERG satellite		
Lunch	Date	2016FY
	Location	Uchinoura Space Center
Configuration	Weight	350kg
Orbit	Altitude	Perigee:about 300km Apogee:about 30,000km
	Inclination	31°
	Type of	Elliptical orbit
	Orbit	
	Period	about 538 min

EXPERIMENTS: The XEP-e's profile shows Table. 2. It has five solid-state silicon detectors (SSDs) and a high-Z scintillator (GSO). The XEP-e is Fig.1. The first SSD discriminates between electron and other particles. And The Other SSDs and a high-Z scintillator decide energy of perticles that are detected.



Fig. 1. XEP-e(Flight Model)

Table. 2.Parformance of the XEP-e		
Energy range	Electron 400keV~20MeV	
G-factor	0.0088 cm ² · sr	
Angular resolution	20° ×22.5°	
Energy resolution	SSDs: less than 20% GSOZ: less than 50%~60%	
Size	317cm × 250cm × 174cm	
Weight	5281g	
Power	19.8W	
Data format	Table mode (18ch) 12ch:SSD SUM 4ch:GSO List mode (for S-WPIA) Quasi-real-time data(for Space Weather)	

We used the KURRI-LINAC (Kyoto University) to calibrate its energy that is between 2MeV and 20MeV electron. We also used our accelerator in Tsukuba space center under 2MeV electrons. We need very low count rate beam like 100~1000cps, we have already got datas to make a beam by using the XEP-e (Engineer Model) last year. In this year we calibrated the XEP-e (Flight Model). As shown in Fig. 2, datas of the the KURRI-LINAC and simulation datas (GEANT4) were very similar between 6MeV and 20MeV. It shows that the KURRI-LINAC can make very low count rate beam.



RESULTS:

The XEP-e could caliblate by using the KURRI- LIN-AC (Kyoto University) that is only a facility in Japan that makes low count rate electrons beam between 5MeV and 20MeV.

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