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- I. Research Reports
- A. Energy Engineering

A.1

Digital Subtraction Cineangiography Using Proton-Induced Dual-Energy Pulsed X-Rays

Yoshiyuki Oguri and Kotaro Kondo

1. Introduction

Digital subtraction angiography is a way of radiography to clearly visualize veins and organs in the patient's body using contrast agent and a computerized X-ray imaging system. The image of the contrast medium can be selectively emphasized by computing the difference between the images taken before and after the injection of the agent. If this technique is applied to cinematography, the diagnostic performance will further be improved. However, it is so far impossible to apply this technique to the diagnostics of fast-moving organs such as cardiac blood vessels, because the injection of the contrast agent requires at least a few seconds.

If we use X-ray photons with an energy slightly above the absorption edge of the contrast agent, the radiographic image of the agent can be highly enhanced. On the other hand, at a photon energy slightly lower than the edge, the image is much weaker, since the absorption by the contrast agent has a minimum. Therefore, we can take a pair of pictures of a blood vessel with different contrasts within a short period by a rapid (≈ 100 Hz) alternation of X-ray energy, instead of alternating the concentration of the contrast medium. Enhanced images of blood vessels are created by subtraction between these two pictures, even if the vessel moves very fast.

High-intensity quasi-monochromatic X-rays can easily be produced by irradiating a pure metallic target with MeV protons[1]. The X-ray photon energy can be changed by changing the target Z number. In this note, the technical feasibility of a high-contrast dual-energy subtraction cineangiography based on proton-induced quasi-monochromatic X-rays is investigated. We present experimental results obtained using a small phantom and a high-sensitivity X-ray movie camera.

2. Experimental method

Figure 1 shows the experimental setup at the tandem accelerator facility at RLNR, Tokyo Tech. Two plates made of different kinds of metallic elements were attached at the diagonal position of a rotating target with respect to the rotation axis. By rotating this target in the vacuum chamber, we could irradiate these two plates alternately by 2.5 MeV protons. Thus quasi-monochromatic pulsed X-rays with two different energies could be alternately produced around the absorption edge of the contrast medium. The duty factor of the X-ray emission was 60%. The repetition frequency was 0-10 Hz. We selected zirconium (₄₀Zr, $hv_{K\alpha} = 15.7$ keV) and niobium (₄₁Nb, $hv_{K\alpha} = 16.6 \text{ keV}$) as the target materials to produce the dual-energy X-rays. The element of the contrast agent was automatically determined to be strontium (₃₈Sr, K-absorption edge energy = 16.1 keV).

The beam current of the incident protons on the target was $0.5-1 \mu A$. X-rays emitted from the targets were extracted through a 50- μ m-thick Mylar window into the atmosphere. The X-ray intensity was measured by a CdTe X-ray detector.



Fig. 1 Experimental setup of the subtraction cineangiography using proton-induced dual-energy pulsed X-rays[2]



Fig. 2 X-ray transmission images of the phantom taken by using the Nb (left) target, Zr (center) target, and the logarithmic difference (right). The parameter α was adjusted so that the background (fish bone + water) on the subtraction image could be minimized[2].

For the movie imaging experiments, we prepared a phantom consisting of a small rubber balloon, water, and a Lucite container. The balloon, which simulated a heart of a small test animal, was filled with aqueous solution of $SrCl_2$ (20%) as the contrast agent. The size of the balloon was ≈ 10 mm in diameter. The motion of the heart was simulated by expanding and contracting the balloon with a piston. A fish (*Pagrus major*) fin bone fragment ($\approx 8 \times 12 \text{ mm}^2 \times 1 \text{ mm}$ thick) was attached to the phantom by adhesive tape in order to simulate a bone around the heart as background. The X-ray transmission images were taken by an X-ray movie camera consisting of a Ce:YAG scintillator and a high-sensitivity low-noise visible-light movie camera.

3. Results and discussion

Transmission images of the phantom taken by using the Nb (left) target, the Zr (center) target, and the logarithmic difference (right) are shown in Fig. 2. In the figure, α denotes a parameter which should be adjusted so that background on the subtraction image could be minimized. An exposure time of 2 s was required for the photography of one frame, owing to low intensity of the X-ray.

Before subtraction, the rubber balloon and the surface of the water can be recognized in both frames (left and center) in Fig. 2. Also, the fish-fin bone simulating the background can be clearly seen on the right-hand side of the balloon. Nevertheless, one sees that the image intensity of the contrast medium in the balloon changes with the X-ray energy. By adjusting the parameter α , the intensity of the images of the fish-fin bone and water in the subtraction photograph (right) could be successfully minimized, and the image of the balloon could be emphasized. However, we see that the image quality was degraded by the digital subtraction, owing to the amplification of the statistical fluctuation of the pixel counts.

We performed a cinematography in a similar manner using the balloon in the phantom being inflated and contracted. A 17-frame digital-subtraction movie was successfully constructed.

4. Conclusions

By using a rotating target and a high-sensitivity X-ray movie camera, we successfully demonstrated a digital subtraction cineangiography based on proton-induced quasi-monochromatic pulsed X-rays.

The attenuation length of the X-rays used in the present research is enough to diagnose small test animals. However, if this technique is applied to actual diagnosis of a human body, the thickness of the object may be a few 10 cm. In this case, much higher X-ray energies (at least several 10 keV) are needed to realize sufficient transmission. For this purpose, a system consisting of a higher-Z contrast agent, higher-Z metallic targets and more energetic incident protons must be developed.

Acknowledgment

The authors wish to thank M. Shikida and S. Matsui for fabrication of the experimental devices. This work was supported by JSPS KAKENHI Grant Number 20300175.

References

- Y. Oguri, J. Hasegawa, M. Ogawa, J. Kaneko, K. Sasa; A Phantom Test of Proton-Induced Dual-Energy X-Ray Angiography Using Iodinated Contrast Media, International Journal of PIXE, Vol. 17, No. 1&2, pp. 11–21 (2007).
- Y. Oguri, Y. Hu, K. Kondo, H. Fukuda, J. Hasegawa; Digital Subtraction Cineangiography Using Proton-Induced Quasi-Monochromatic Pulsed X-Rays, International Journal of PIXE, Vol. 23, No. 1&2, pp. 21–29 (2013).

A.2 Framing images of discharge plasmas in the electromagnetic shock tube for ion-beam-atomic hydrogen interaction experiments

Kotaro Kondo and Yoshiyuki Oguri

1. Introduction

Energy deposition from heavy ion beams to a target is important information to understand the physical state of the final target condition for heavy-ion-fusion and heavy-ion-driven high energy density physics experiments. Moderate energy ions such as ~ MeV/u with a sub-range target can be utilized for high energy density physics experiments. The ions can give the energy to the sub-range target efficiently and the energy deposition depends on the electronic-state of the target. Since there is no experimental data of the stopping power with the dissociation effect of target molecule, which is one of the basic chemical effects, experimental investigation is of particular interest. The hydrogen was chosen as the atomic target because a hydrogen molecule has only valence electron.

An electromagnetic shock tube worked as a driver for generation of well-defined dissociated targets. The interaction between the discharge current and the induced magnetic field accelerates the gas and causes the shock wave formation. The shock tube generated a quasi-steady and one-dimensional strong shock wave [1]. The shock-wave method has a strong point that a physical state of matter behind the shock can be estimated by the Rankine-Hugoniot relations from the shock velocity. However, the previous shock velocity was not enough to dissociate the hydrogen completely. We performed experiments on the framing images of the discharge plasma in the shock tube to understand the shock wave physics and to improve the shock velocity.

2. Experimental setup

The experimental setup is shown in Fig. 1. The shock tube was filled with hydrogen gas with pressure from 400 to 1000 Pa. The hydrogen gas was supplied from a hydrogen generator (Horiba Stec, Inc., OPGU-2200). The shock tube had a coaxial structure. A gap switch was connected between capacitors ($3.5 \ \mu$ F) and the central electrode with a 12-mm outer diameter. The inner diameter of the outer electrode was 30 mm. The charging voltage was 17 kV. The main discharge occurred just after the gap switch received a trigger signal. We measured the discharge voltage by a high voltage probe (Tektronix, Inc., P6015A) [2].

The high-speed framing camera, which consisted of two CCD cameras (Hamamatsu, Inc., C6558) and high-speed gated image intensifier units (Hamamatsu, Inc., C2925-01) gave two pictures for the single discharge. Each exposure time was 10 ns. The camera focused on the coaxial electrode section, which had a slit window with a 5-mm width. The operation of the camera was synchronized with the pulse discharge.



Fig. 1 Experimental setup including the electromagnetic shock tube and the high-speed camera.

3. Experimental results

The measured waveform is shown in Fig. 2. We assume the electrical circuit of the experimental setup as an RLC circuit. The voltage V(t) as a function of time is given as

$$V(t) = V_0 \exp\left(-\frac{R}{2L}t\right) \left(\cos\sqrt{\frac{1}{LC} - \frac{R}{4L^2}}t + \frac{R/2L}{\sqrt{1/LC - R^2/4L^2}} \sin\sqrt{\frac{1}{LC} - \frac{R}{4L^2}t}\right),$$

where V_0 , *L*, *C*, *R* are the charging voltage, inductance, capacitance, and resistance, respectively. Fitting the measured waveform with this equation by the method of least squares showed L = 270 nH, R = 50 m Ω . *C* was the total capacitance of 3.5 µF and V_0 was the initial charging voltage of 17 kV in these experimental conditions. The voltage was fully damped at $t \sim 30$ µs. The waveform did not depend on the initial pressure.



Fig. 2 Discharge waveform. Solid and dash lines mean the experimental data and fitted curve based on the RLC circuit, respectively.

Using the high-speed framing camera, we obtained self-emission profiles of the discharge plasma shown in Fig. 3. The left picture of Fig. 3 shows vertical discharge plasmas.

These experimental results indicate that the discharge plasma was not radially homogeneous. Such an inhomogeneity can cause insufficient acceleration from the interaction between the discharge current and the induced magnetic field, which may results in a low shock velocity.

4. Summary

Framing images of the discharge plasma in the electromagnetic shock tube were successfully obtained by the high-speed framing camera.

These experimental results showed the vertical discharge plasma and may cause insufficient acceleration for the hydrogen gas.

Acknowledgement

We would like to thank S. Matsui for preparation of the experimental devices.



Fig. 3 Framing images of the discharge plasma in the electromagnetic shock tube taken by the high-speed framing camera. The initial hydrogen pressure was 1000 Pa. The discharge occurred at t = 0.

References

- Kotaro Kondo, Mitsuo Nakajima, Toru Kawamura, and Kazuhiko Horioka: "Compact pulse power device for generation of one-dimensional strong shock waves", Review of Scientific Instruments 77, No.3, pp. 036104-1-3, 2006.
- [2] Kotaro Kondo, Jun Hasegawa, Kazuhiko Horioka, and Yoshiyuki Oguri: "Velocity measurement of electromagnetically-driven shock wave to produce a dissociated hydrogen target for beam interaction experiment", Energy Procedia, in press.

A.3 Spectroscopic Examination of γ–band Spectra of NO Molecule by Using Microwave Discharge N₂-O₂ Mixture Plasma

Hiroshi Akatsuka, Atsushi Nezu, Haruaki Matsuura

Nitrogen oxides (NO_x) have been a hot topic in the last decades from the viewpoint of remediation of atmospheric environment due to their toxicity. Monitoring and removal process requires fundamental data on NO molecule, such as molecular constants or spectroscopic characteristics, particularly, of its excited states. Since NO plasma has strong radiation lines as a couple of band spectra even with a low electron density, it is easy to observe them by the OES method.

The γ -band radiation is originated from the electronic transition from NO A $^{2}\Sigma$ metastable state to X $^{2}\Pi$ ground state, which is shown in Fig. 1. We carried out a theoretical calculation of the γ -band radiation. By using the optical emission spectroscopic measurement method, we experimentally examined the γ -band spectrum emitted from microwave discharge N₂-O₂ mixture plasma. Then, we fit the observed spectrum with our calculation as shown in Fig. 2, so that the vibrational and rotational temperatures of NO A $^{2}\Sigma$ state were obtained.

The N₂-O₂ plasma is generated by a microwave discharge with its frequency 2.45 GHz, and the output power is set at 400 W. The total pressure of the plasma ranges from 0.5 to 2 Torr. A CCD spectrometer (200 to 850 nm) is applied as a polychromator for the measurement of the emission spectra.

From the experimental results, it is found that both NO and N_2 molecules in the mixture plasma experience cooling down processes as the flowing distance increases both on vibrational and rotational temperatures. On the other hand, NO molecules have always the higher rotational temperature than N_2 , and the rotational damping process is slower than that of the vibration. It implies that the rotational excitation kinetics of NO molecule is different from N_2 . As we know, NO molecules in $N_2 - O_2$ mixture



Fig. 1 An example of spectra radiated from microwave discharge $N_2 - O_2$ mixture plasma with its pressure about 1 Torr in the present study. NO γ -band and N_2 second positive system (2PS) are observed. The spectral sensitivity is not calibrated.



Fig. 2 Fitting example of NO γ -band spectral results using an N₂ – O₂ = 1:1 discharge plasma with 1 Torr pressure (experiment: dots, calculation: hollow rectangle, $\Delta v = -3$, FWHM = 2 nm).

plasma come from collisions among a variety of molecules and atoms.

Another remarkable finding in this study is as follows. When O₂ molar ratio of the mixture increases, the NO experiences an increasing vibrational temperature monotonically, while N2 2PS shows non-monotonic variation in its vibrational temperature, which is shown in Fig. 3. This is because that the NO A $^{2}\Sigma$ metastable state is excited from two main paths: one is the collision between NO X $^{2}\Pi$ with free electrons and the other is the collision between NO X $^{2}\Pi$ and N₂ A $^{3}\Sigma$. These two ways have different vibrational excitation processes, and the collision between NO X $^{2}\Pi$ and N₂ A³ Σ appears a vibrational energy exchange. Hence, when the participator density changes, the NO A $^{2}\Sigma$ vibration property should depend on a molar ratio of O₂ [1−3].



Fig. 3 Vibrational temperature of NO A $^{2}\Sigma$ state (solid dots) and N₂ C $^{3}\Sigma$ state (hollow triangle) plotted against partial pressure of oxygen in a plasma feeding gas.

Reference

1. H. Tan, A. Nezu, H. Matsuura and H. Akatsuka: Proc. ICRP8/SPP31, 5B-PM-O5, Fukuoka, Japan (2014).

2. H. Tan, A. Nezu, H. Matsuura and H. Akatsuka: Bull. Am. Phys. Soc., **59** [16], p. 19 (2014).

3. H. Tan, A. Nezu, H. Matsuura and H. Akatsuka; Jpn. J. Appl. Phys., **54** [1S], 01AB06 (2015).

A.4 Hybrid Numerical Simulation of Supersonic Flow of Weakly-Ionized Rarefied Arc-Jet along Diverging Magnetic Field

Hiroshi Akatsuka, Atsushi Nezu, Haruaki Matsuura

We have been experimentally examining physical properties of expanding arc-jet plasma [1]. Upon our previous study, we must carry out numerical simulation of the expanding plasma jet as rarefied gas dynamics based not on fluid approximation but on particle modeling, which is precisely the objective of this study. We carried out hybrid simulation of weakly ionized rarefied helium arc-jet plasma flowing supersonically along open magnetic field lines to understand the mechanisms of acceleration and deceleration of supersonic plasma and of electric potential formation. We considered background residual helium molecules in the plasma expansion region. In the simulation, ions and neutrals were treated with DSMC simulation since the Knudsen number of the system was in the order of ~ 0.01 - 1. On the other hand, electrons were treated by fluid model in order that the spatial discretization should become reasonable.

The results are summarized in Fig. 1. Thus, the number densities of any species were found to decrease by an order of magnitude as the plasma flowed to the downstream direction till the distance from the magnet exit $z \sim 0.15$ m. After that, they were found to turn into increase till the end of the domain considered $z \sim 0.30$ m. On the other hand, as for the velocity change, it was found that the maximum Mach number of ions and neutrals are about 2 at z = 0.12 m downstream direction from the exit of the hollow magnet. After that, the plasma flow including neutrals was decelerated into subsonic velocity. Meanwhile, the plasma temperature as well as neutral one became the lowest around the maximum velocity position, and after that, they increased, which was interpreted that collisional relaxation of plasma became dominant in this area, i.e., the kinetic energy of fluids was converted into thermal energy.

Another interesting finding was the velocity difference between charged particles and neutrals, i.e., a kind of ion slip. Ions and electrons were accelerated till $z \le 0.12$ m. The present simulation showed that the total space potential dropped almost monotonically by ~ 1.0 V from the exit of the magnet to the end of the calculated domain. However, the mechanism of the potential drop was found to be rather complicated. That is, until about the maximum velocity position $z \sim 0.15$ m, the pressure drop made the space potential lower. On the other hand, the plasma-slip turned to decrease remarkably in the downstream region, and therefore, the potential drop by the friction effect became essential in the most downstream region [2 – 3].

Reference

1. K. Yoshida, T. Shibata, A. Nezu, H. Matsuura and H, Akatsuka, IEEE Trans. Plasma Sci., **37**, 1414-1418 (2009).

2. H. Akatsuka, S. Tsuno, A. Laosunthara, A. Nezu, and H. Matsuura, Bull. Am. Phys. Soc., **59**, [16], 75 (2014).



Fig. 1 2-D maps of the (a) magnetic field calculated as input conditions, (b) ion density contour n_i , (c) vector diagram of the ion velocity v_i , (d) ion temperature contour T_i , (e) contour map of electric potential ϕ , (f) neutral density contour n_n , (g) vector diagram of the neutral velocity v_n , and (h) neutral temperature contour T_n , respectively. In the figures, parameters with a superscript * indicates their values at z = 0.

3. S. Tsuno, T. Nakahagi, R. Yamashiro, A. Nezu, H. Matsuura, H. Akatsuka, IEEE Trans. Plasma Sci., **42**, [12], 3732-3741 (2014).

A.5 Flowing Characteristics of Cold Nitrogen Arc Jet Plasma along Open-Field-Line

Hiroshi Akatsuka, Atsushi Nezu, Haruaki Matsuura

Plasma expansion along an open-field-line has various applications in active engineering research fields, such as space propulsion and material processing, since it allows control of ion fluxes. Up to the present time, we investigated such plasma-flow characteristics of helium [1] and argon plasmas [2] flowing supersonically along open-field line. In this study as a next step, we experimentally measure the ion Mach number and plasma parameters of cold nitrogen plasma with an electron temperature of less than 1 eV along an open-field-line. Nitrogen plasmas are practically crucial since it can be applied as a nitrogen radical source for hardening of metallic materials. We already examined its spectral characteristics and found remarkable peculiarity, i.e., strong atomic nitrogen lines and weak molecular band spectra [3]. In the present study, we examined characteristics of expanding nitrogen plasma, particularly of its fluid-dynamic characteristics, which gives essential fluid dynamic information of nitrogen plasma jet as a thruster for astronautic applications for artificial satellites. The plasma jet generator applied in the present study is the same with our previous studies. Its detailed account was already described elsewhere [1 - 3]. To generate the nitrogen plasma stably, we added argon gas into the discharge gas as buffer medium, whose partial pressure ratio is set as $\text{He:N}_2 = 4:0, 3:1, 2:2, \text{ and } 1:3$. Typical discharge conditions are as follows: pressure of the discharge region $1 \times 10^4 - 2 \times 10^4$ Pa, that of the plasma



Fig. 1. Ion Mach number measured by a para-perp Mach probe on the plasma jet axis r = 0. The partial pressure ratio of the feeding gas was set as He:N₂ = 4:0 in (a) and 1:3 in (b).



Fig. 2. Electron temperature on the plasma jet axis r = 0. The partial pressure ratio of the feeding gas was set as $\text{He:}N_2 = 4:0$ in (a) and 1:3 in (b).

expanding region 0.1 Pa, arc discharge current 120 A, arc voltage about 17 - 25 V. We measured the electron temperature, density and plasma potential by a Langmuir probe for, while the Mach number by a para-perp Mach probe.

Figure 1 is the axial profile of the ion Mach number, where (a) and (b) correspond to the mixture ratio He:N_2 = 4:0 and 1:3, respectively. As was shown in the figure, it is found that the maximum position of the Mach number gradually shifts monotonically to the downstream direction as the partial pressure of nitrogen increases. Meanwhile, Fig. 2 shows the axial profile of the electron temperature for the same discharge conditions. It is found that the temperature lowering continues farther in the downstream area for high-N₂ concentration, whereas the pure helium discharge makes the temperature-lowering limit in the upstream area. We are now discussing this difference in the plasma parameters according to the chemical species of the discharge gases.

Reference

1. K. Yoshida, T. Shibata, A. Nezu, H. Matsuura and H. Akatsuka; IEEE Trans. Plasma Sci., **37**, [8], 1414-1418 (2009).

- 2. Y. Nagahara, H. Ichii, K. Yoshida, A. Nezu, H. Matsuura and H. Akatsuka; IEEE Trans. Plasma Sci., **41**, [8], 1869-1877 (2013).
- 3. H. Akatsuka, M. Hatcho and H. Matsuura; IEEE Trans. Plasma Sci., **42**, [12], 3691-3697 (2014).

A.6 Synthesis of super ionic conductive ceramics using molten salt flux and elucidation of their electric conduction mechanism: EXAFS analysis of various rare earth oxyfluorides at high temperature

Haruaki MATSUURA, Atsushi NEZU and Hiroshi AKATSUKA

To cultivate novel application of high temperature molten salt from the way to electrolysis or thermal conductive media, production of amorphous ionic conductance material has been considered. At the first step, lanthanum oxyfluoride which exhibits ionic conductance charasteristics at high temperature is chosen and the relationship between local structure around lanthanum and ionic conductance has been investigated. In this year, local structure around lanthanum lanthanum fluoride, of lanthanum oxyfluoride and their mixture with lithium fluoride depending on temperature has been evaluated by extended X-ray absorption fine structure.

Each component properly weighed was mixed with boron nitride powder in a few microns, and pressed into pellets in 13 mm diameter and 0.5 mm thickness. The mixing weight ratio of chemicals to BN was ca. 1 to 2.5. These pressed pellets were installed inbetween pyrolytic boron nitride plates which havw been developed by the CEMHTI, CNRS, France [1-3]. To prevent oxidation of sample during heating process in EXAFS mesurement, these pellets were heated in a gas tight electric furnace filled with He gas at ca. 30 kPa. The La L_{III}-edge EXAFS spectra have been collected with fixed time scan method by the X-ray from a double Si (111) crystals monochromator in transmission mode at BL27B/PF/KEK, Japan. EXAFS data were analysed by using the WinXAS ver.3.1.

Figure 1 shows the temperature dependence of the interionic distance between La3+ and anion of various materials. By the investigation of local structure around lanthanum depending on temperature, interionic distance between La³⁺ and F⁻ expands at 700 °C, and super ionic transition temperature decreases from bulk material by the effect of mixing with boron nitride matrix. Thus, mixing with matrix can make stabilization of super ionic conductance phase. In the case of lanthanum oxyfluoride, at 300 °C, interionic distance between La and anion once shortens and expands even more rapidly than the case of LaF₃. Thus, it would also be advantageous of keeping super ionic conductance characteristics. To insert the material among boron nitride powder, it's essential to samples being in molten, thus lithium fluoride was chosen in this study. In the case of lanthanum fluoride and lithium fluoride in 1 to 1 composition, which doesn't have any chemical stoichiometric stable compounds, interionic distance between La3+ and anion the most extends among the samples tested. Thus, melting with matrix material makes super ionic conductance phase stabilized further. In the next step, thermodynamics of

chemicals and conductance of heat treated materials will be carried out systematically.

This study was financially supported by a preliminary research grant of RLNR, Tokyo Tech.



Fig. 1 Interionic distance between La^{3+} and anion depending on temperature.

References

 C. Bessada, et al, Nucl. Sci. NEA/NSC/DOC 15 (2009) 117.
M. Numakura, et al, J. Fluor. Chem. 131, (2010) 1039.
M. Numakura, Doctor Thesis, Tokyo Institute of Technology (2011).

A.7 Pyrochemical treatment of fuel debris using selective fluorination and molten salt electrolysis: Fluorination behavior of zirconium oxide

Haruaki MATSUURA, Atsushi NEZU and Hiroshi AKATSUKA

How to take out and stabilize fuel debris from the Fukushima Daiichi power plant is the one of imporant processes to be developed for decomission. We propose one of pyrochemical processes by the combination of selective fluorination and molten salt electrolysis in order to reduce the secondary wastes. Historically, fluorination reaction of actinide oxides has been relatively investigated in the course of development of fluorine volatilization technique, but that of zirconium oxide is apparently lacking. For the first stage of development, the fluorination of zirconium oxide has been performed at various temperatures by using fluorine or hydrofluoride gas.

Fluorination reactor (Figure 1) using fluorine or hydrofluoride gas was constructed in a drought chamber at the hot laboratory in the IMRAM, Tohoku University. To avoid from the hydration of fluorinated samples, all samples after treated by fluorination were handled in an Ar circulated glove box. Both sample chamber and sample pans were made from pure nickel metal. The sample fluorinated has been characterized by the powder X-ray diffraction.

First, fluorination of pure zirconium oxide using fluorine gas at various temperatures for several durations has been performed. By introduction for only 1 hour of He+F₂(5%) 20 ml/min at 700 °C, the weight of product decreases, thus vaporization as ZrF₄ would be expected. The most effective condition for avoiding vaporization is at 500 °C for 2 hours. The X-ray diffraction pattern of product is shown in Fig. 2(a). 90% of products contains α ZrF₄ and small amount of β ZrF₄ and oxyfluoride were identified too.

Next. fluorination of zirconium oxide using hydrofluoride gas has been performed. Even by the introduction of 20 ml/min + Ar 30 ml/min at 600 °C for 2 hours, the weight decreases. Fluorination rate using hydrofluoride gas is less than that using fluorine gas. As shown in Fig.2(b), only 30% was converted to αZrF_4 and plenty amount of ZrO₂ was left. In real debris, zirconium oxide would be existing with uranium oxide as solid solution. If fluorine gas is used for fluorination agent, UF_6 is expected to be vaporized at 300 °C. Thus, hydrofluoride gas is suitable to be used for fluorination agent in order to avoid vaporization.

Fluorination reaction of uranium oxide and solid solution by hydrofluoride gas is now under investigation [1,2].

This study was done by a collaboration research with Prof. N. Sato, Tohoku University, and financially supported by JSPS KAKENHI, Grant Number 25630429.



Fig. 1 Fluorination equipment constructed at IMRAM, Tohoku University.



Fig. 2 X-ray diffraction patterns (Cu K α) of fluorination products of ZrO₂ by (a) F₂ and (b) HF gases at 500 °C for 2 h.

References

[1] H. Matsuura, *et al*, The 2nd Asian Nuclear Fuel Conference, 2014, Sendai, Japan, 029.

- Conference, 2014, Sendar, Japan, 029.
- [2] H. Matsuura, et al, The Nuclear Materials Conference,
- 2014, Clearwater beach, USA, O709.

A.8 Structural analysis of cations in molten sub-halide systems: Fluoride addition effect on UV-vis spectra of neodymium cation in molten chlorides

Haruaki MATSUURA, Atsushi NEZU and Hiroshi AKATSUKA

Neodymium is the one of the rare earth fission products and was utilized as the prototype of some trans uranium elements due to its similarity of electrochemical behavior. Also, neodymium is one of important materials since it has been widely utilized at magnets in the motors, and effective recycling technology from the end produncts has been waiting for a long time. Although molten salt should be relatively constructed by a simple structural model due to the predominant ionic species in liquid phase, several electrochemical behaviors have not been well understood by the microscopic point of view. For a recent few years, the electrochemical behavior of rare earths including neodymium in the molten chlorides with small amount of fluorides has been focused, and structural elucidation by extended absorption fine structure and UV-vis spectroscopy of neodymium in molten salts has been performed [1,2]. In this year, fluoride concentration dependence on the spectra of neodymium in molten NaCl-CsCl-NaF was measured more precisely than previous year and the structural variation is discussed by some parameters derived from the Judd-Ofelt analysis.

UV-vis spectroscopy of neodymium in molten NaCl-CsCl-NaF at 953 K has been carried out by using the spectrophotometer (JASCO V-500) in KURRI. A quartz cell with 10 mm of light path was used for the molten salt container, and experimental procedure was exactly the same as that described in the report of the last year [3]. For the calculation of oscillator strength of the hypersensitive transition and derivation of $\Omega_{2,4,6}$ parameters by the Judd-Ofelt analysis, density was assumed to be the additivity of molar volume of each component, and refractive index was temporary used from the value of KCl due to the non-availability of the data of CsCl.

Figure 1 shows the variation of oscillator strength of hypersensitive transition absorption of neodymium at ca. 589 nm depending on fluoride addition. This absorption peak has been considered to the indication of coordination symmetry around neodymium cation, i.e. 6 coordinated octahedral species, and the smaller value relates to more perfectly symmetric. One striking feature is that with increasing fluoride amount until ca. F/Nd = 2 in molten NaCl-CsCl-NaF, the value of oscillator strength once increases until F/Nd = 2, and decreases rapidly. While in molten LiCl-KCl-LiF, the value of oscillator strength decreases almost linearly.

Figure 2 shows $\Omega_{2,4}$ values derived by the Judd-Ofelt analysis. Ω_2 value is in the trend of decreasing with increasing F/Nd ratio, but at ca. F/Nd = 3 in NaCl-CsCl-NaF and at F/Nd = 6 in LiCl-KCl-LiF, small hump appears. Smaller value of Ω_2 is considered to relate covalency of local structure around neodymium cation, thus the melt around F/Nd = 3 in NaCl-CsCl-NaF and around F/Nd = 6 in LiCl-KCl-LiF may take un-stabilized structure, in which local coordinated chlorides are half exchanged by fluorides. Ω_4 , which is suggested to relate the donation effect of electrons from coordinated anions, seems to show much complicated tendency depending on F/Nd ratio, thus further systematic investigation on the similar mixture systems would be interested.

This study was done by a collaboration research with Profs. T. Fujii, A.Uehara and H. Yamana, Kyoto University.



Fig. 1 Oscillator strengths of the electronic absorption spectra of neodymium in molten LiCl-KCl-LiF and NaCl-CsCl-NaF.



Fig. 2 $\Omega_{2,4}$'s of the electronic absorption spectra of neodymium in molten LiCl-KCl-LiF and NaCl-2CsCl-NaF.

References

[1] Y. Shimohara *et al*, Molten salt chemistry and technology, John Wiley & Sons (2014) 577.

- [2] K. Fujita et al, Proc. AMS4, (2012) 264.
- [3] K. Fujita, Master thesis, Tokyo Tech. (2013).

A.9

Hydration Behavior of Mg(OH)₂-Ni(OH)₂ Composite Prepared by Mechano-chemical Method

Junichi RYU and Yukitaka KATO

Introduction

In recent years, the technologies of chemical heat pump and chemical heat storage are widely studied for the reduction of energy consumption and CO_2 emission. Chemical heat pump system with the reaction between magnesium oxide (MgO) and water vapor has been reported by our group. [1]

$$MgO + H_2O \rightleftharpoons Mg(OH)_2 \quad \Delta H = -81.2 \text{ kJ mol}^{-1}$$

In this system, thermal energy above 350° C is required for practical heat storage operation. Recently, chemically modified magnesium hydroxides were proposed as new type material for chemical heat storage by our group. These materials can store thermal energy around 280 °C. [2-4] The heat storage density of these materials are higher than authentic magnesium hydroxide under 280°C of heat storage condition. In this work, hydration behaviour of Mg(OH)₂-Ni(OH)₂ composites prepared by mechanochemical method as a new material for chemical heat storage were studied by TG method.

Experimental

 $Mg(OH)_2$ -Ni(OH)_2 composite was prepared by mechano-chemical method. Authentic $Mg(OH)_2$ and Ni(OH)_2 were used as precursor. Mixture of $Mg(OH)_2$ and Ni(OH)_2 with several molar ratio were milled in planetary ball mill under 400rpm of rotation speed. Solid state reaction with milling treatment was confirmed by FT-IR. Hydration reaction of the prepared samples at 110°C with 57.8 kPa of water vapor were tested by thermogravimetric method after dehydration at 300°C. Hydration conversion of samples were calculated from the weight change of sample.

Results

Hydration behavior of Mg-Ni (9:1) prepared by mechano-chemical method and Mg_{0.9}Ni_{0.1}(OH)₂ prepared by co-precipitation method under the reaction temperature of 110°C and vapor pressure of 57.8 kPa, after dehydration at 300°C for 30 min, are shown in Fig. 1. The values of hydration conversion, ΔX_1 , for Mg-Ni (9:1) and Mg_{0.9}Ni_{0.1}(OH)₂ were 39.5% and 16.3%, respectively. This indicates that the hydration reactivity of Mg-Ni (9:1) was higher than Mg_{0.9}Ni_{0.1}(OH)₂. The SEM image of Mg-



Figure 1 Hydration behavior of Mg-Ni (9:1) and $Mg_{0.9}Ni_{0.1}(OH)_2$.



Figure 2 SEM image of Mg-Ni (9:1) prepared by mechano-chemical method.

Ni (9:1) is shown in Fig. 2. The particle size of Mg-Ni (9:1) was smaller than 10 μ m, and that was quite smaller than Mg_{0.9}Ni_{0.1}(OH)₂ (5 μ m ~ 50 μ m). These results suggest that the difference of hydration reactivity for Mg-Ni (9:1) and Mg_{0.9}Ni_{0.1}(OH)₂ shown in Figures 1 was due to the difference of particle size of these samples.

The stability for cyclic operations and heat output performance under practical conditions should be studied further.

References

- 1 Y. Kato et al., Appl. Therm. Eng., 16, 852 (1996).
- 2 J. Ryu et al., J. Chem. Eng. Jpn., 40, 1281 (2007).
- 3 H. Ishitobi et al., *Ind. Eng. Chem. Res.*, **52**, 5321 (2013).
- 4 J. Ryu et al., J. Chem. Eng. Jpn., 47, 579 (2007).

A.10 Development of an atmospheric-pressure plasma source through cooperation with a technical high school

Noriyosu Hayshizaki

1. Introduction

In the national curriculum guidelines for high school teaching, "project study" is defined as an industrial course subject. According to the subject commentary, "project study is the subject which aims to train a problem-solving ability and creative attitude through the learning to deepen acquired knowledge and skill", and it corresponds to creativity development courses of Tokyo Tech.

Although technical high schools are one of the educational institutions for developing human resources in manufacturing in our country, it is said that there are few chances for technical high school students to meet scientists and actually learn about advanced science and technology. In order to expand opportunities in this area, a new approach to organic cooperation between technical high schools and universities was attempted by using an atmospheric-pressure plasma source as educational materials.

2. Development of atmospheric-pressure plasma source

Development of an atmospheric-pressure plasma source is experimental subject of university-level. Its system which is compact and easy to handle provides safety plasma observation for high school students. Graduate students carried out visiting lectures about a particle accelerator, a plasma source and their own studies in "project study" at Komagane Technical High School in Nagano Prefecture. Under advice by the staff of Tokyo Tech, technical high school students fabricated the atmospheric-pressure plasma source by themselves and succeeded in plasma production. After the completion of the experiments, they made a summarized report and gave an oral presentation at the first Japan national conference on cooperation between technical high schools and universities.



Fig. 1 Visiting lectures by graduate students



Fig. 2 Collaborative experiment



Fig. 3 The atmospheric-pressure plasma source



Fig. 4 Presentation by technical high school students

Acknowledgment

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B.1 Progress of Neutron Capture Cross Section Measurements

at Tokyo Institute of Technology

Masayuki Igashira, Tatsuya Katabuchi, Kazushi Terada, Brian Hales, Shotaro Yanagida, Takuro Arai

1. Introduction

The neutron capture reaction is important in nuclear engineering, astrophysics and fundamental nuclear physics. In recent years, nuclear transmutation systems to transmute long-lived fission products (LLFP) and minor actinides (MA) in nuclear waste to stable or shorter-lifetime isotopes are becoming an attractive option for long-term management of nuclear waste. To develop a nuclear transmutation system, accurate neutron capture cross section data of those radioactive isotopes are required. In other fields, neutron capture cross sections are particularly important inputs for stellar nucleosynthesis models.

In 2013 fiscal year, our Tokyo Tech research group carried out experiments to determine neutron capture cross sections for both fields, nuclear engineering and stellar nucleosynthesis. In addition, a project for medical application is ongoing in our group. Two neutron facilities, a Pelletron accelerator of the Tokyo Tech and the Japan Proton Accelerator Research Complex (J-PARC) in the Japan Atomic Energy Agency, were used for the experiments.

2. Tokyo Tech

2.1. Barium-138

We measured the neutron capture cross section and the neutron capture γ -ray spectrum of ¹³⁸Ba in the keV neutron energy region. Bariuim-138 has a small neutron capture cross section because of closed neutron shell structure of N=82. In the slow neutron capture process (*s*-process) in stellar nucleosynthesis, ¹³⁸Ba acts as a bottleneck of the *s*-process path. Accurate neutron capture cross section of ¹³⁸Ba is required to calculate *s*-process production yields in stellar nucleosynthesis models.

Experiments were performed using a neutron beam from the ${}^{7}Li(p,n){}^{7}Be$ reaction induced by a proton beam from the Pelleton accelerator. The incident neutron energy distributed from a few keV to 100 keV. The neutron energy was determined by the time-of-flight (TOF) method. The incident neutron spectrum was measured with a ⁶Li glass scintillator located at a distance of 30 cm from the neutron source. An isotopically enriched ¹³⁸Ba sample was used. The sample was placed at a neutron flight distance of 12 cm. Gamma-rays emitted from the neutron capture reaction were detected with an anti-Compton NaI(Tl) spectrometer. The pulse-height weighting technique was applied to derive neutron capture yields. Runs for a gold sample were also made as reference measurements to the standard cross section of the 197 Au(n, γ) 198 Au reaction.

2.2. Online dosimetry system for boron neutron capture therapy

This research is for developing an online dosimetry system for the boron neutron capture therapy (BNCT). BNCT uses high LET particles from the ${}^{10}B(n,\alpha)^7Li$ reaction to terminate cancer cells. Cancer cells can be selectively killed with boron-contained compounds taken in cancer cells. However there is one issue to transform BNCT to a widespread radiation therapy. Currently, absorbed dose to tumor and normal tissues of a patient cannot be measured during the irradiation. To solve the issue, we are developing an online dose imaging system for BNCT [1,2], in which 478 keV γ-rays from the ${}^{10}B(n,\alpha)^7Li^*$ reaction are detected through collimators and the distribution of ${}^{10}B(n,\alpha)^7Li$ reaction rate is reconstructed from the projection images of detected counts. We built a test version of the system and performed test experiments using a neutron beam from the ${}^{7}Li(p,n){}^{7}Be$ reaction. A cylindrical water phantom having two boron-concentrated regions was irradiated with the neutron beam. The two high reaction rate regions were successfully separated in a reconstructed image.

3. J-PARC

3.1. Development of pulse-width analysis method

A spallation neutron source of J-PARC gives a pulsed intense neutron beam with a wide range of energy. The Accurate Neutron Nucleus Reaction Measurement Instrument (ANNRI) was built on the No. 4 neutron beam line of J-PARC for neutron capture cross section measurements [3,4]. The strong neutron beam of J-PARC facilitates new experiments that could not be made before but a fast signal processing system for data acquisition is required. In particular, a strong γ -ray burst called gamma flash occurs every time the 3-GeV proton beam strikes the spallation target. The gamma flash makes a large input to the detector, thus causing a saturated output, and shifting and ringing of the baseline. Signals cannot be processed properly due to the baseline distortion for a certain duration of time after the gamma flash. A detector system needs to quickly recover from the saturated condition by the gamma flash.

We have developed a new signal processing method based on pulse-width analysis for neutron capture cross section measurements using an NaI(Tl) detector of ANNRI [5]. The pulse width of the anode output signal from the NaI(Tl) detector was measured with a time digitizer. Analog modules such as amplifiers for pulse-height analysis are not necessary. This makes the system more robust to the gamma flash. We tested the method in ANNRI. Pulse-height spectra were successfully reconstructed from pulse-width spectra using the conversion line determined from γ -ray sources and neutron-induced reactions.

3.2. Palladium-107

The neutron capture cross section of ¹⁰⁷Pd, one of the LLFPs produced in nuclear power plants, was measured using the NaI(Tl) detector of ANNRI. The pulse-widh method described above was employed for data acquisition. The incident neutron spectrum was determined with the ¹⁰B(n, α)⁷Li reaction. The pulse-height weighting tecnique was applied for deriving neutron capture yields. The cross section was determined from 10 meV to 100 keV. The results were normailized to the cross section value of JENDL-4.0 at a resonance peak of 44 eV. The results were described in Ref. [6].

3.2. Americium-241

Americium-241 is contained in spent nuclear fuel and dominates the radioactivity of nuclear waste for long time. Thus, ²⁴¹Am is in the high priority list for nuclear transmutation. The neutron reaction data of ²⁴¹Am is very important for design of nuclear transmutation system. We measured the neutron capture cross section of ²⁴¹Am using

the NaI(Tl) detector of ANNRI. The experimental method was the same as ¹⁰⁷Pd described above.

Acknowledgment

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Reference

1. B. Hales et al.; Applied Radiation and Isotopes, Vol.88, pp. 167-170 (2014).

2. T. Katabuchi et al.; Applied Radiation and Isotopes, Vol.88, pp. 139-142 (2014).

3. M. Igashira et al.; Nuclear Instruments and Methods in Physics Research Section A, Vol.600, p.332 (2009).

4. Y. Kiyanagi, et al.; Journal of Korean Physical Society, Vol.59, p.1781, (2011).

5. T. Katabuchi et al.; Nuclear Instruments and Methods in Physics Research Section A, Vol.764, pp. 369-377 (2014).

6. K. Terada et al.; Progress in Nuclear Energy, in press.

B.2 Liquid-Liquid Extraction of Uranyl Species Using Ionic Liquid, Betainium Bis(trifluoromethylsulfonyl)imide

Kotoe Sasaki, Tomoya Suzuki, Takahiro Mori, Tsuyoshi Arai, Koichiro Takao, Yasuhisa Ikeda

1. Introduction

Recently, functionalized ionic liquids (ILs), so-called task-specific ILs (TSILs) have been synthesized and applied in various fields. In the nuclear industry field, TSILs are expected to be used as media for radioactive waste treatments and the reprocessing of spent nuclear fuels. In fact, the dissolution properties of uranium oxides (UO_2, U_3O_8) into ILs, the extraction behavior of uranyl species from aqueous solutions to IL or TSIL phases, electrochemical properties of uranyl species in ILs, and properties of some other actinoid species in ILs have been studied extensively.

We have also studied dissolution behavior of uranium oxides (UO₂ and U₃O₈) and electrochemical properties of uranyl species in BMI (1-butyl-3-methylimidazolium) based ILs ([BMI]Cl, [BMI]BF₄, and [BMI][NfO] (NfO: nonafluorobutanesulfonate)) to examine the feasibility as media for pyro-reprocessing processes for spent nuclear fuels [1-3]. As a result, it was clarified that UO₂²⁺ (U(VI)) species in [BMI]Cl are almost reversibly reduced to UO₂⁺ and that UO₂²⁺ species in [BMI][NfO] are reduced to U(IV) with formation of deposits on the electrode.

Binnemans et al. have reported that betainium bis(trifluoromethylsulfonyl)imide ([Hbet][Tf₂N]) can dissolve selectively metal oxides such as Sc_2O_3 , Y_2O_3 , Ln_2O_3 (Ln = La, Nd, Sm, Eu, Gd, Dy, Ho, Er, Tm, Yb, Lu), UO₃, ZnO, and its binary mixture with water forms one homogeneous phase more than 60 °C, i.e., thermomorphic behavior [4-7].

Based on our previous studies and the reports of Binnemans et al., we have proposed that [Hbet][Tf₂N] should be used as the medium in decontamination processes of wastes contaminated uranium such as NaF and Al_2O_3 adsorbents generated from the ²³⁵U enrichment facility.

As a basic study for developing the decontamination method using [Hbet][Tf₂N],we examined the extraction behavior of U(VI) species from aqueous HNO₃ solutions to [Hbet][Tf₂N] phase, Similarly, the extraction behavior of Na(I) and Al(III), which are main components of NaF and Al₂O₃ adsorbents, was also examined.

2. Experimental

The dried [Hbet][Tf₂N], which exists as a solid at room temperature, was prepared using the literature method [4]. [TMPA][Tf₂N](TMPA:N,N,N-trimethyl-N-propylammonium) was synthesized by stirring H₂O-CH₃CN mixture dissolved [TMPA]Br and LiTf₂N with a 1:1 molar ratio for 12 h at room temperature. [Hbet][Tf₂N] and $[TMPA][Tf_2N]$ used in this study were pre-equilibrated by contact with aqueous HNO₃ solutions of various concentrations. The pre-equilibrated aqueous and IL phases were separated each other. Into the resulting aqueous HNO₃ solutions. $UO_2(NO_3).6H_2O_1$ NaNO₃. or Al(NO₃)₃·9H₂O was dissolved to prepare sample solutions of 2.0×10^{-2} M (M = mol/dm³), respectively. The obtained aqueous HNO₃ solutions (2 ml) were loaded in test tubes together with the pre-equilibrated [Hbet][Tf₂N] or $[TMPA][Tf_2N]$ (2 ml), followed by vigorous shaking in a thermostat bath for 1 h at 25°C. After the centrifugal phase separation, the concentrations of M^{n+} (U(VI), Al(III), or Na(I)) species in the aqueous HNO₃ phases were determined by ICP-AES. The extractability (E, %) and distribution ratio (D) were calculated by the following equations, respectively.

$$E = 100 \times ([\mathbf{M}^{n+}]_{\text{ini}} - [\mathbf{M}^{n+}]_{\text{aq}})/[\mathbf{M}^{n+}]_{\text{ini}}$$
(1)

$$D = ([\mathbf{M}^{n+}]_{ini} - [\mathbf{M}^{n+}]_{aq})/[\mathbf{M}^{n+}]_{aq}$$
(2)

where $[M^{n+}]_{ini}$ and $[M^{n+}]_{aq}$ denote the M^{n+} concentrations in aqueous HNO₃ phases at the initial state and after the extraction, respectively.

3. Results and Discussion

Firstly, we examined the time for attaining extraction equilibrium in the extraction of U(VI) from 0.01 M HNO₃ aqueous solution to [Hbet][Tf₂N] phase. As a result, the extraction equilibrium was found to be attained sufficiently by almost 7 min. Hence, all extraction experiments in this study were performed in the shaking time of 1 h.

In order to examine the effects of acid concentration and the cationic components of ILs on the extraction of U(VI), we performed the extraction experiments from aqueous HNO3 solutions of various concentrations (0.01, 0.1, 0.3, 0.5, 1.0, 2.0 M) to ILs phases ([Hbet][Tf₂N] or [TMPA][Tf₂N]). The obtained E values are plotted against [HNO₃] as shown in Fig. 1. As seen from this figure, the Evalues decrease with an increase in [HNO₃] in [Hbet][Tf_2N] system. This suggests that the U(VI) extraction is due to the complex formation of UO_2^{2+} through the COO⁻ group formed with deprotonation of [Hbet]⁺ (bet), because its deprotonation should be depressed with increasing [HNO₃]. In fact, by using pK_a value of $[Hbet]^+$ (1.83), the mole fractions of bet in 0.01 and 1.0 M HNO₃ solutions are estimated to be 0.60 and 0.015, respectively. This is considered to be a reason for a

lowering of E values of U(VI) in high acidic solutions. The complex formation of UO_2^{2+} with bet in low acidic solutions is supported by facts that the $[(UO_2)_2(bet)_6 (H_2O)_2$ [Tf₂N]₄ complex has been isolated as the single crystals, that the six bet molecules coordinate to the equatorial plane of uranyl moiety through COO⁻ oxygen, and that the U(VI) species exist as $[UO_2(bet)_3]^{2+}$ in the [Hbet][Tf₂N] solution dissolved $[(UO_2)_2(bet)_6(H_2O)_2]$ $[Tf_2N]_4$ [7]. On the other hand, the U(VI) species were little extracted in $[TMPA][Tf_2N]$, in which $[TMPA]^+$ has not COOH group. This result also supports that the extraction of U(VI) to [Hbet][Tf $_2N$] phase is attributed to the complex formation of UO_2^{2+} with bet. The D value corresponding to the E value (62 %) in the extraction from 0.01 M HNO₃ solution is evaluated to be 1.6, which is larger than that (D < 0.1) in the extraction of U(VI) from 0.01 M HNO₃ aqueous solution to 30% tributyl phosphate/dodecane phase. This suggests that the [Hbet][Tf_2N] itself is the relatively effective extractant for U(VI) species.



Fig. 1. Plots of *E* values vs. $[HNO_3]$ for the extraction of U(VI) from HNO₃ solutions to $[Hbet][Tf_2N]$ or $[TMPA][Tf_2N]$ phase.

Furthermore, in order to investigate the applicability of [Hbet][Tf₂N] to the decontamination of wastes contaminated uranium such as NaF and Al₂O₃ adsorbents, we examined the extraction behaviour of Na(I) and Al(III), which are main components of NaF and Al₂O₃ adsorbents used in the ²³⁵U enrichment processes. The plots of *E* values vs. [HNO₃] for Na(I) and Al(III) species are shown in Fig. 2 with that for U(VI). As seen from this figure, the Na(I) and Al(III) species are not extracted from the aqueous HNO₃ solutions to [Hbet][Tf₂N] phase under the present conditions. This result suggests that in the low HNO₃ concentration range the U(VI) species are separated selectively from Na(I) and Al(III) species.

From the present study, it is expected that the decontamination method for the uranium wastes using [Hbet][Tf₂N] should be feasible, that is, the leaching of uranium components from the wastes by soaking uranium wastes into the homogeneous solution of water and [Hbet][Tf₂N] formed at temperature more than 60 °C, and

then the extraction of only U(VI) species into [Hbet][Tf₂N] phase with the phase separation by cooling to the room temperature.



Fig. 2. Plots of *E* values vs. [HNO₃] for the extraction of $U(VI)[\circ]$, Na(I)[Δ], and Al(III)[\Box] from HNO₃ solutions to [Hbet][Tf₂N] phase.

Reference

1. N. Asanuma, et al., Korea-Japan Workshop on Nuclear Pyroprocessing, Kyongju, Korea, Abstr. 30, 2006.

2. N. Asanuma et al., 2nd COE-INES International Symposium INES-2, Yokohama, Japan, Abstr. 55, 2006.

- 3. N. Asanuma et al., J. Nucl. Sci. Technol., 44, 368-372 (2007).
- 4. P. Nockemann et al., J. Phys. Chem. B, 110, 20978-20992 (2006).
- 5. P. Nockemann et al., Inorg. Chem., 47, 9987-9999 (2008).
- 6. P. Nockemann et al., Cryst. Grouth Des., 8, 1353-1363 (2008).
- 7. P. Nockemann et al., Inorg. Chem., 49, 3351-3360 (2010).

B.3 Après ORIENT - Separation Adsorption Characteristics of Fission Products in High Level Liquid Wastes on Macro-cyclic Resins

Masaki Ozawa, Toshitaka Kaneshiki and Chie Iguchi

1. Introduction

Après ORIENT research program, newly initiated in 2011, will deal with transmutation of radioactive fission product elements to create rare metals / rare earth (RE) elements by (n,γ) reaction with subsequent β - decays in the reactors. We call this reaction as modern alchemical reaction,

 $_{Z}\text{FP}^{A}(n,\gamma) _{Z}\text{FP}^{A+1} \rightarrow _{Z+1}\text{NRM}^{A+1} + \beta^{-},$

where target $_{Z}FP^{A}$ must be radioactive, and eventually created $_{Z+1}NRM^{A+1}$ should be more valuable, strategic and non or extremely less radioactive than that of the target.

2. Separability

In the context, separation of $_{Z}FP^{A}$ from high level liquid wastes (HLLW) for target fabrication, and mutual separation of $_{Z}FP^{A}$ and $_{Z+1}NRM^{A+1}$ will be required after the transmutation. Fig.1 shows the resin polymerized in the micro pores of silica carrier, <u>ca</u>. 60µm. Functional crown ether (CE) are Benzo-15-crown-5, Benzo-18-crown-6 and Dibenzo-21-crown-7.



Fig.1 Polyphenol-based Thermosetting Resin

Fig.2-i,ii,iii show the typical adsorption characteristics of the element for Benzo-18-Crown-6 (BC18) -substituted resin in the simulated HLLW / 5M HCl medium.



Fig.2(i) Relation Between D for Macro-cyclic B18C6 Resin and Ionic radii(Å) of ions in $5\underline{M}$ HCl-simulated High Level Liquid Waste Solution



Fig.2(ii) Relation Between D for Macro-cyclic B18C6 Resin and Ionic radii(Å) of Trivalent ions in 5MHCl-simulated High Level Liquid Waste Solution



Fig.2(iii) Relation Between D for Macro-cyclic B18C6 Resin and Surface Charge Density (C/r^2) of ions in 5<u>M</u>HCl-simulated High Level Liquid Waste Solution

Fig.3-i,ii show the typical adsorption characteristics of the element for Benzo-18-Crown-6 (BC18) -substituted resin in the simulated HLLW / 5M HNO₃ medium.



Fig.3(i) Relation Between D for Macro-cyclic B18C6 Resin and Ionic diameter(Å) of ions in $5\underline{M}NO_3\text{-simulated}$ High Level Liquid Waste Solution



Fig.3(ii) Relation Between D for Macro-cyclic B18C6 Resin and Surface Charge Density (C/r^2) of ions in 5<u>M</u>HNO₃-simulated High Level Liquid Waste Solution

As a conclusion, newly synthesized macro-cyclic resins, *e.g.*, B18C6, must be a vital tool for such separation. Following results were newly obtained.

• In HCl media, the ions with larger diameter with lower surface charge density suggested higher affinity (D) to B18C6 resin. Such tendency was more distinct in HCl media, especially among mono-, di-and tetravalent ions.

• Highest distribution ratio(D) of Ba^{2+} in 5<u>M</u>HCl media would be explained by its lowest surface charge density with specific contribution like "Size fitting Effect" by crown ether ring of 18C6 resin.

• No affinity of REE^{+3} and $PGM(Ru^{3+}, Rh^{3+})$ with B18C6 resins in either HNO₃ or HCl medium.

• Also, such bulky, oxoanionic ions as MoO₄²⁻, ReO₄⁻,

and probably For TcO_4^- suggested rather high affinity in HCl medium.

• In combinatorial chromatographic separation, mutual separation of PGM (Ru, Rh, Pd) would be probable; total sptn. of PGM from HLLW by tertiary pyridine resin, succeeded by selective adsorption of Pd²⁺ by B18C6 macro-cyclic resin.

Rare Metals are inevitable for leading industries, and thus hold the national GDP. Industry-oriented **PGM** and **RE**, etc., are highly strategic, fate-decision material like **OIL**. FP should be considered as not simply radioactive waste, but artificial resource ore. *Après ORIENT*, enhanced recycling of FPs, will contribute either to decrease radiotoxicities and amounts of wastes, or conversely to increase new, non radioactive resources.

References

1.Masaki Ozawa, Toshitaka Kaneshiki, Thomas Sychra, Chie Iguchi and Shin Okumura, Adsorption Characteristics of Fission Product Elements Constituting High Level Liquid Wastes for Macro-cyclic Resins, *ISEC2014 International Solvent Extraction Conference*, 07-11 September 2014, Congress Center Wurzburg (CCW), Germany.

B.4

Après ORIENT - Transmutation **Rhenium Production from Tungsten in Fast Reactors**

Masaki Ozawa and Atsunori Terashima

1. Introduction

Après ORIENT research program, newly initiated in 2011, engages the transmutation of fission product (FP) elements to create, stable or very short-lived rare metals / rare earth (RE) elements by (n,γ) reaction with subsequent β^{-} decays in the reactors.

 $_{Z}FP^{A}(n,\gamma) _{Z}FP^{A+1} \rightarrow _{Z+1}RM^{A+1} + \beta^{-}$ As for its application, Rhenium (Re, VII group element) production by irradiating Tungsten (W) metal was evaluated. Re is one of the rarest metals, and W-Re is well-known, heat-resistant super-alloy for jet-engine, for instance.

2. Transmutation

In the Fig. 1, upper data of each horizontal line indicates the neutron energy averaged capture cross section in fast reactors while the lower does that in light water reactors (LWRs). The differences are one order or more, therefore the exact evaluation of neutron energy spectra of the reactors or zones of the reactor where W is irradiated is crucial to determine the production rate of Re. Although the conventional ORIGEN-2 with one group cross section system^{*2} is used for calculating the transmutation of each nuclides, the continuous energy Monte Carlo Code $\ensuremath{\mathsf{MVP}}^{*3}\ensuremath{\mathsf{was}}$ employed, where the neutron energy spectra of each zone of reactors are exactly simulated, and detailed configurations for evaluating the effective neutron cross sections and neutron flux was well evaluated.

The total flux of fast reactor (FR) is generally one order higher than that of LWRs, thus a middle scale fast reactor is assumed to employ for irradiating the target assembly that contains W pellets in the pins.

Fig.2 illustrates the core layout and target assemblies for Re production from W in the fast reactor with 710 MWt. A total of 54 target assemblies containing inner 19 pins of tungsten (W) pellets surrounded by 42 pins of zirconium hydride(ZrH₂) pellets are set at the location of the peripheral of the core, the equivalent diameter and height of which is about 1.8 m and 0.9 m, respectively. Hydride is employed to increase the transmutation rate by slowing down the fast neutrons, where the capture cross section of W is larger at lower neutron energy regions as shown in Fig.3..











Fig.1 Transmutation scheme of Re production from irradiated W in reactors

The change of the isotopes during two years including one year cooling is shown in Fig.4. Fig.4 indicates that Re-188, the main radioactive istope, will be rapidly decayed if Re is separeted from W just after the irradiation. In 4 months cooling, the secondly intensive radioactive Re-186 is decreased less than the exemption level, 1000 Bq/g.



Fig.4 Change of isotopes during irradiation and cooling

Table 1 summarized the transmutation of W. Case 1 of Table 1 lists the weight of the transmutation from natural W to total Re after one year irradiation. Case 2 of Table 1 indicates the Re production when neutron moderator is not employed, and where the whole 61 pins of the target assembly are W metal. The loaded weight of W in Case 2 is 4 times more than that of Case 1, although the production of Re is one order less than that of Case 1. This is mainly due to the neutron spectrum softening effect by zirconium hydride.

Table 1 Total mass of W and Re along with irradiation and cooling

Case	Number of W pins	Number of ZrH ₂ pins	Loaded weight of W(g)	Produced weight of Re after one year irradiation(g)
1 (Reference)	19	42	1.6×10 ⁶	1.4×10^{4}
2(No moderator)	61	0	5.0×10^{6}	1.5×10 ³

The effective capture cross section of W-186, the main parent of Re, is 1.731 barn for Case 1 evaluated by MVP code, while that of Case 2 is 0.012 barn. This cross sections of Case 2 seems much smaller than 0.597 barn, because the former is the effective one considering the heterogeneous effect on neutron self shielding and the latter is that for infinitely diluted nuclide. Precise seaparation of Re from rad. W, Ta and Os will be issue.

Reference

1) Tsugio Yokoyama, Atsunori Terashima and Masaki Ozawa, Evaluation of Rhenium Production Rates in Tungsten Irradiated in Fast Reactors by Using Continuous Energy Monte Carlo Code MVP, *The 8th International Symposium on Technetium and Rhenium: Science and Utilization(ISTR2014)*, Proc.pp.526-532, September 29th to October 3rd 2014, La Baule - Pornichet, France

B.5 Synthesis of Crown-Ether Resins and their Strontium-Selectivity

Toshitaka Kaneshiki and Masaki Ozawa

1. Introduction

After the catastrophe of Fukushima daiichi NPP on March 11, 2011, the valid processing method of radioactive strontium becomes necessary. The purpose of this study is development of practical and inexpensive strontiumselective resin. Some crown-ethers form the complex with strontium, therefore crown-ether resin can be adsorbent to select the strontium. Three crown-ether resins were synthesized in silica beads, and evaluated strontiumselectivity.

2. Experiments & Results

2.1. Synthesis of the resins

Crown-ether, bisphenol A and paraformaldehyde were dissolved in trichloroacetic acid (TCA). This solution was polymerized at 90°C, in porous silica beads with 5 μ m diameter (Fig.1). And crown-ethers (CE) which used reaction were shown in Fig.2.



Fig. 1 Reaction formula of resins

Name	Formula	Hole Diameter	CE content/resin
Benzo−15− crown−5 (BC15)		170-220 pm	0.397 mmol/g
Benzo−18− crown−6 (BC18)		260-320 pm	0.382 mmol/g
Dibenzo-21- crown-7 (DBC21)		340-430 pm	0.354 mmol/g
4,4'(5')-Di-t-butyl- cyclohexano-18- crown-6 (Sr Resin)		260-320 pm	

Fig. 2 Three kind of crown-ethers (CE) & Sr Resin

2.2. Adsorption experiment (Batch test)

Synthesized 3 CE-resins and Sr Resin of Eichrom Tech. Inc. were mixed with 0.1M-Sr solution which was prepared for 0.1-9M-HCl. After 24-hour-shaking at 25 $^{\circ}$ C, Sr concentration of each solutions were measured with ICP-MS and calculated adsorption percentage and distribution coefficient (Kd). A result was shown in Fig.3.





2.3. Adsorption experiment (Column Chromatography)

 $0.8 \text{cm} \phi \text{ x30 cm}$ glass column was filled 7.4g of BC18 resin. Various cations of Na, K, Mg, Ca, Sr and Ba(each concentration was 0.5mmol/dm³ in 9M-HCl) wrer adsorbed in this column. Adsorbed cations were eluted by 9-0.1M-HCl. Chromatogram was shown in Fig.4.



Fig. 4 Chromatogram of various cations separation,

3. Conclusions

BC18 resin showed high Sr selectivity in high concentration of HCl. Results of column chromatography showed that this resin was possible to separate Sr from other cations in seawater, too. We want to use this resin as Sr adsorption agents in the future.

Acknowledgment

This study was the result of the joint research with Engineering Research & Development Center of ATOX CO.,LTD.

Reference

1. T. Sakurai, et al.; ATOX TECHNICAL REPORT, No.5, pp. 10-11(2013) (in Japanese).

2. M.Ozawa. et. al : Global 2013, Salt Lake City, UT (United States), 29 Sep - 3 Oct (2013).

B.6 Heavy Metal Freezing Behavior for Stop of Water Leakage from LWR Containment Vessel

Minoru Takahashi, Mihail Mihalache, Radu Secareanu, Alexandre Chapelat

1. Introduction

In the decommissioning of Fukushima Daiichi Nuclear Power Plants, the reactor pressure vessels (RPV) and the reactor containment vessels (RCV) must be filled with water to shield radiation before the removal of core debris from the RPV. It can be achieved by stopping water leakage from the RCV. Concrete has been the candidate of sealing material. However, liquid heavy metals such as lead alloys solidify around the leakage position in water atmosphere more quickly than the concrete. They can be radiation shield, and are stable under radiation condition for the decommissioning period of 30-40 years. In the present study, the applicability of liquid heavy metal to the stop of the leakage is investigated.

The promising candidate of the liquid heavy metal may be lead alloys, particularly Wood's metal (48%Bi-26%Pb-13%Sn-13%Cd) with a melting point of 72°C. The melting point less than 100°C is required for the freezing of the metal without disturbance of boiling under atmospheric pressure.

The freezing phenomenon of the liquid heavy metal is important for stopping the water leakage from objects of complex shapes. The present study proves that heavy metal can be used as a water leak plug material. An experimental installation was developed to investigate the behavior of the stop of water leak and metal freezing using Wood's metal and simulant metal with low melting point: gallium. The results of this study can lead to the development of the method of the stop of water leakage that can be used for the decommissioning of nuclear reactors after accidents.

2. Experiments

2.1. Liquid metal discharge and freezing test

2.1.1 Experimental apparatus

In order to investigate the freezing capabilities of liquid metal and demonstrate that it was appropriate for stopping the water leak, an experimental installation was developed for discharge test using gallium and Wood's metal. Gallium was chosen as a simulant metal for preliminary experiment because it is easy to do the experiment using the metal with much lower melting point. The physical properties of the metals are shown in Table 1 in comparison with those of lead-bismuth eutectic. Lead-bismuth eutectic was not chosen because its melting point of 124.5°C caused water boiling at atmospheric pressure. In case of occurrence of water boiling, the liquid metal will be disturbed by steam bubbles during its freezing, and as a result the shapes of frozen surfaces become rugged.

The discharge and freezing experiment was conducted using water as coolant. A schematic diagram of the experimental apparatus is shown in Fig. 1. The facility consists of the liquid metal tank, the vertical cylinder with level meters, the test section, the water circulation loop and the plate cooling water loop. The water in the test section leaked through a hole of the metal plate, and flowed back to the test section through the pump at desired temperature. The metal plate at the bottom was cooled by the water in the plate cooling water loop. The levels of liquid metal in the left supply tank and in the right cylindrical tank with the level meters were balanced. Then, the horizontal pipe between the tanks was plugged by freezing the liquid metal inside by water-cooling. The cover gas in the cylindrical tank was pressurized by nitrogen gas to discharge the liquid metal downward to the test section through a tube.



Fig. 1 A schematic diagram of experimental apparatus

2.1.2. Test section

The rectangular test section with transparent side windows was used as shown in Fig. 2. The metal plate was located horizontally on the bottom of the test section. The atmosphere above the metal plate was air or water. The water entered from the side wall and flowed out through the hole of the metal plate. The water temperature inside of the tank was controlled by using an electric heater. Liquid metal was discharged downward from the nozzle made of the tube with an inner diameter of 4.4 mm. The water temperature and the nozzle outlet temperature were measured by thermocouples.

In order to investigate the freezing phenomenon on the metal plate, a hole was made in the center of the plate to simulate a water leak, and four thin thermocouples were inserted near the hole from the bottom to measure the temperature distribution. Two thermocouples were located 1 mm and the other two 5 mm in depth from the top surface to record the temperature history during freezing of liquid metal. In order to see the effect of the metal plate material on freezing behavior, two types of metal plates were used: one made of stainless steel 304 and another made of brass

(60%Cu-40%Zn).



Fig. 2 Test section

2.1.3. Experimental conditions

Wood's metal temperature was in the range of 80-100°C which was higher than the melting point of the liquid metal and lower than the boiling point of water at the atmospheric pressure. Two cases of plate cooling and no plate cooling were chosen as a test condition, where the plate cooling simulated the condition of a cooled structure around leak position by cold water before liquid metal discharge. The experiment was conducted with both water and with air atmospheres in the test section.

2.2. Experimental test of freezing of static Wood's metal

In order to create a simplified simulation model of metal freezing and verify it by means of comparison of calculated result with experimental data, static freezing experiment was conducted using the test section shown in Fig. 3. The cylindrical cup made of brass was immersed in a water pool and hot liquid Wood's metal was poured into the vessel. The temperatures at the top and bottom surfaces of the bottom wall of the cup were measured using thin thermocouples during the cooling and freezing of the Wood's metal.



Fig.3. Test section for static freezing of Wood's metal

3. Result

3.1. Preliminary discharge and freezing test with gallium

Fig. 4 shows the appearance of the frozen gallium over the hole on a brass plate (yellow color). The frozen gallium was smooth on the upper surface, and the projected rod was formed on the lower side by liquid gallium penetration into the hole. The water temperature was kept constant, and the gallium mass flow rate was about 40 g/s. The temperature of gallium changed from 24 °C to about 45 °C.



Fig. 4 Frozen gallium over hole on brass plate

Fig. 5 shows the result of gallium penetration length into the hole determined from the length of the solid rod. The penetration length is larger with the increase of the diameter of the hole, since liquid gallium freezes worse in larger holes. It is smaller in case of plate cooled and particularly in the water atmosphere. It means the cooling of the solid plate around a leak hole is effective for metal freezing and stopping of water leakage.



Fig.5 Penetration length of gallium in preliminary test

3.2. Discharge and freezing test with Wood's metal

Fig. 6 shows the appearance of frozen Wood's metal on the plates of brass and stainless steel, and the debris that were formed after the liquid Wood's metal flowed through the hole and froze. The frozen metal spread over the surface more widely on the stainless steel plate than on the brass plate. It means that the Wood's metal did not freeze soon and spread more on the stainless steel plate because coolability by the stainless steel plate was worse than that by the brass plate. The amount of debris is more in the brass plate. This may be caused by the fact that the liquid Wood's metal was cooled faster by the brass plate that had higher thermal conductivity than by the stainless steel plate, and that it froze without spreading more over the plate, and the melt went through the hole of the brass plate more without being cooled than the stainless steel plate.



Fig.6 Frozen Wood's metal.

Almost all the Wood's metal froze on the plate, but some fraction of the melt entered into the hole and flowed down through the hole. The melt that flowed down became debris below the test section. The fraction of the debris formed in the total amount of discharged Wood's metal is shown in Fig. 8. The fraction of debris is lower with lower water temperature and lower Wood's metal temperature, in particular in case of plate cooling. It means that the freezing is enhanced by these conditions. Therefore, these conditions is better for stopping water leakage.



Fig.7 Penetration length of Wood's metal



Fig.8 Debris of Wood's metal

3.3. Static freezing test with Wood's metal

Fig. 9 shows an example of the comparison of the measured and calculated temperatures at the top and bottom surfaces of the bottom wall of the brass cup. It can be seen that the calculated result shows the same trend with the measured result. This means that the present one-dimensional freezing model is adequate for extention to the simulation of the discharge and freezing behavior.



Fig.9 Measured and calculated temperature in static Wood's metal freezing

4. Conclusion

The fundamental experiment was conducted using gallium and Wood's metal to investigate the discharge and freezing behavior for development of technical method of stop of water leakage. It has been found that the freezing feature is better in cases of the heavy metal discharged to a water-cooled metal plate with higher thermal conductivity at lower heavy metal temperature in a water atmosphere rather than an air atmosphere. The one-dimensional unsteady heat conduction and freezing model could predict the temperature in static freezing of Wood's metal, which suggests the capability of simulation of the practical freezing phenomena of liquid metal for stop of water leakage with the extention of this model. The present result is useful to develop an equipment for stopping the water leakage in a water environment using heavy metal freezing.

Reference

1. Minoru Takahashi, Mihail Mihalache, Radu Secareanu, Alexandre Chapelat, "Fundamental Study on Heavy Metal Freezing Behavior for Stop of Water Leakage from LWR Containment Vessel," 4th International Symposium on Innovative nuclear Energy Systems (INES-4), Tokyo, November 6-8, 2013.
B.7 Development of Thin-film Coating Technique of TPPEN-NIPA Gel in Silica Particles for MA/Ln Extraction Separation

Kenji TAKESHITA, Yusuke INABA, Hiroshi OAKI and Hideharu TAKAHASHI

In HLLW (High-level Liquid Wastes), some fission products and these daughter nuclides $({}^{90}\text{Sr}/{}^{90}\text{Y}$ and ¹³⁷Cs/^{137m}Ba) cause the radioactivity for the first 300 years and after that some minor actinides (MA, 241 Am, 243 Am/ 239 Np and 244 Cm) cause the radioactivity. If partitioning of MAs from HLLW and transmuting of MAs by fast reactors are realized, the radiotoxicity of HLLW can be reduced for a short period. In this partitioning, we are developing a MA/Ln extraction separation process using chromatographic agents produced by innovative polymer technologies such as the chemical immobilization of functional ligand into polymer gel and the thin-film coating of the polymer gel on the pore surface of porous medium ⁽¹⁻⁶⁾. The rapid extraction and elution of metal ion are required for the establishment of a chromatographic separation process with polymer gel. A polymer technology to coat thin-film of polymer gel on the pore surface of porous silica is indispensable to increase the extraction and elution rates. In this report, we introduce the thin-film coating technique of TPPEN-NIPA(N,N,N',N'-tetrakis(4-propenyloxy-2-pyridyl methyl)ethylenediamine-N-isopropylacrylamide) gel on

methyl)ethylenediamine-N-isopropylacrylamide) gel on porous silica particles. In our coating process, porous silica particles with the average pore diameter of 300 nm and 50 nm are prepared. And a monomer mixture of NIPA and TPPEN are diluted with DMF (N,N-dimethylform- amide), in which a small amount of AIBN (azobisisobutyronitrile) is added as a polymerization initiator, is immersed in these porous silica particles by capillary force. The TPPEN-NIPA gel is produced by the radical polymerization technique.

Fig.1 shows the SEM observation of the porous particles before and after the coating of the gels with the TPPEN content of 3%. It should be noted that most pores were remained after the gel coating, although a part of small pores were plugged by the gel. Thickness of the gel on the pore surface was changed by the temperature rising rate for the radical polymerization. Thin-film gel can be formed on the pore surface of the porous silica particles by the faster temperature rising rate above 5°C/min. The radical polymerization started at the temperature around 70°C and proceeded spontaneously and promptly by the generation of polymerization heat. Both the thickness and the content of the gel immobilized in the porous silica particles decreased with increasing the TPPEN content (Fig.2). For the porous silica particles with the average pore diameter of 300nm, the average thickness of the gel was evaluated to be in the range of 15 to 38nm. For the silica particles with small pores of 50nm in diameter, the average thickness of the gel became thinner and was in the range of 2 to 5nm.

In conclusion, we succeeded in the thin-film coating of TPPEN-NIPA gel on silica particles and clarified the

optimum coating condition in which thin film of NIPA-TPPEN gel with the average thickness of 2 to 40 nm was formed on the pore surface in porous silica particles by the radical polymerization with fast temperature rising rate.



Fig.1 SEM observation of porous silica before and after gel coating (Q300: average pore diameter 300nm, Q50: average pore diameter 50nm)



Fig.2 Relation between the average thickness of the gel and the gel content

Reference

- 1. K. Takeshita, T. Ogata, H. Oaki, Y. Inaba, A. Mori, T. Yaita and S. Koyama, *GLOBAL 2013 Int. Nuclear Fuel Cycle Conf.*, 554-558 (2013).
- 2. K. Takeshita, Y. Inaba, T. Yaita, S. Suzuki, A. Mori, *SCEJ 45th Autumn Meeting*, N123 (2013).
- 3. K. Takeshita, H. Oaki, T. Yaita, S. Suzuki, A. Mori, *SCEJ 45th Autumn Meeting*, N124 (2013).
- 4. K. Takeshita, Y. Inaba, T. Yaita, S. Suzuki, A. Mori, *Proc. 32nd Symp. Solv. Extr.*, B-12 (2013).
- 5. K. Takeshita, H. Oaki, T. Yaita, S. Suzuki, A. Mori, *Proc. 32nd Symp. Solv. Extr.*, B-13 (2013).

6. K. Takeshita, H. Oaki, T. Yaita, S. Suzuki, A. Mori, 247th ACS National Meeting & Exposion, 115 (2014).

B.8 Physical property changes of polymorphs of silicon nitride and SiAlON ceramics by neutron irradiation

Toyohiko YANO, Areerak Rueanngoen and Katsumi Yoshida

Silicon nitride and related ceramics are excellent engineering ceramics and of interest for nuclear applications because of their excellent mechanical and other properties. Silicon nitride and related ceramics include the wide range of compounds, which are categorized into two basic crystal structures, α - and β -Si₃N₄ structures. Reports discussing neutron irradiation effects on SiAlON ceramics are very limited, and almost no data is available on difference in polymorphs. In this research, α and β phase of Si₃N₄ and SiAlON were prepared by hot-pressing to evaluate neutron irradiation effects. Si₃N₄ and SiAlON ceramics were neutronirradiated in the Japan Materials Testing Reactor up to a fluence of 8.5x10²⁴ n/m² (E>0.1 MeV) at 563 K. Changes in macroscopic length and lattice parameter due to the irradiation were measured. Changes in macroscopic length of α - and β -SiAlON and α -Si₃N₄ were similar magnitude $(0.11 \sim 0.14\%)$, and almost double that of β -Si₃N₄ (0.06%). The lattice parameter changes of SiAlONs showed a complicated trend, expansion of the a-axis and contraction of the c-axis in both phases. No obvious microcracking, void formations or loop formations were observed in SiAlONs microstructures[1].

1. Introduction

Ceramic materials are attractive for fusion applications because of their excellent thermal and mechanical properties, such as high thermal shock resistance and high mechanical strength. From the beginning of the material development for fusion applications, ceramic materials such as Al_2O_3 , $MgAl_2O_4$, Si_3N_4 , MgO, SiAlONs and SiChave been expected to be suitable materials for fusion applications. The ceramics with more complex crystal structures and stoichiometries, such as $MgAl_2O_4$, Si_3N_4 and silicon oxynitride, appear to resist voids-induced expansion better than simple oxides. The excellent strength and thermal shock resistance of Si_3N_4 and SiAlONs suggested that they are expected to be candidate materials for fusion application.

Basically, radiation effects are influenced by crystal structures of compounds; α -Al₂O₃ and AlN showed larger swelling and degradation of thermal diffusivity than β -Si₃N₄ and β -SiC after the irradiation more than 1x10²⁶ n/m² [2]. In addition, macroscopic length expansion of Si₃N₄ was roughly one-half of that of SiC after the same irradiation conditions (2.8-4.2x10²⁶ n/m², 753-1003 K) [3].

High-resolution transmission electron microscope (HREM) observations of heavily neutron-irradiated Si_3N_4 revealed the presence of dislocation loops laid parallel to the c-axis [4,5]. Furthermore, HREM observation of irradiated β -Si₃N₄ after post-irradiation annealing showed bubbles of He generated by transmutation of ¹⁴N [6].

In this study, changes in macroscopic length and lattice parameters of silicon nitride and SiAION ceramics were measured after moderate fluence irradiation up to 8.5×10^{24} n/m² (E>0.1 MeV) at 563 K to clarify basic irradiation effects on each polymorph. In addition, microstructural defects in these specimens were observed by HREM.

2. Experimental Procedures

Silicon nitride and SiAlON ceramics were prepared by hot-pressing technique from raw powder mixtures. These powders were mixed using a ball mill for 24 h with ethanol as a medium. Dried powder mixtures were hot-pressed under a uniaxial pressure of 30 MPa. SEM observation indicated that grain shape of α -Si₃N₄ was equi-axed, and the grain size distributed between 100 and 500 nm (250±80 nm in average). Beta-Si₃N₄ showed the mixture of large elongated grains with the aspect ratio about 4-9 and small equi-axed grains. Elongated-grains preferentially grew ranging from 100 to 800 nm in diameter and 500 nm to 4 µm in length. Average grain size of small equi-axed grains was 440±190 nm. Average grain sizes of α - and β -SiAlON were 330±180 and 800 ±150 nm, respectively. Grain morphologies of both SiAlONs were equi-axed. Both phases of Si₃N₄ contained relatively large amount of grain boundary glassy phases, while both phases of SiAlON did not contain thick grain boundary phases.

The sintered samples were cut into the rectangular bars of 25 mm in length and 2x4 mm² in cross section, then they were concurrently neutron irradiated in the Japan Materials Testing Reactor of a fluence up to 8.5×10^{24} n/m² (E>0.1 MeV) at 563 K for 1209 h in an He-filled capsule. Macroscopic length of four specimens of each material was measured before and after the irradiation using a point-type micrometer at room temperature. The accuracy of the length measurement was ±1 µm or 0.004 % of 25mm sample. Lattice parameters were observed by XRD technique. XRD with a CuK α source was performed on a Philips PW-1700 diffractometer equipped with a graphite monochrometer at room temperature (299±1 K). The Xray diffractometry was done using Si powder as an

Specimen	Macroscopic length change (%)	Macroscopic volume change (%)	Lattice parame	Unit-cell volume	
			a-axis	c-axis	change (%)
α -Si ₃ N ₄	0.11[1]	0.33[3]	0.0619[2]	0.0897[2]	0.2137[3]
β -Si ₃ N ₄	0.06[1]	0.18[3]	0.0341[1]	0.0399[2]	0.1081[3]
α-SiAlON	0.12[1]	0.36[3]	0.0822[5]	-0.051[1]	0.114[1]
β-SiAlON	0.14[1]	0.42[3]	0.1321[5]	-0.109[1]	0.155[2]

Table 1 Changes in macroscopic length and lattice parameters of neutron irradiated-Si₃N₄ and SiAlON ceramics. after irradiation of 8.5×10^{24} n/m² at 563 K.

internal standard (a_0 = 0.5430758 nm at 298.1 K), which was stuck on the specimen surface with glue.

Microstructure of irradiated and unirradiated specimens was investigated by HREM. The HREM used in the present study was Hitachi H-9000 high resolution TEM operated with an accelerating voltage of 300 kV. The thin foils for HREM were prepared by dimpling and ionmilling method.

3. Results and Discussion

Changes in macroscopic lengths and lattice parameters of Si₃N₄ and SiAlON ceramics after neutron irradiation are summarized in Table 1. Changes in the macroscopic length of α - and β -Si₃N₄ and α - and β -SiAlON were 0.11, 0.06, 0.12, and 0.14%, respectively. Changes in the macroscopic lengths of SiAlON ceramics and α -Si₃N₄ were similar magnitude (0.11~0.14%) and almost double that of β -Si₃N₄. The lattice parameter expansion in both axes of α-Si₃N₄ was larger than those of β -Si₃N₄; however, it was slightly smaller compared with their macroscopic length change. The lattice parameter changes in SiAlON ceramics showed a complicated trend. The a-axis parameter slightly expanded, while the c-axis parameter shrank slightly in both α - and β -SiAlON. The unit cell volume expansions of α - and β -Si₃N₄ were 0.2137 and 0.1081% and those of α - and β -SiAlON were 0.114 and 0.155%, respectively, while their macroscopic volume changes were 0.33, 0.18, 0.36, and 0.42%, respectively.

Table 2 shows FWHM of XRD peaks of the unirradiated and irradiated specimens. The FWHM of peaks after the neutron irradiation increased. It indicated that the internal stain may increased after the neutron irradiation.

Table 2 Full width at half maximum of XRD peaks of unirradiated and irradiated specimens.

Spaaiman	Indices -	FWHM [deg]		
specifien		Unirradiated	Irradiated	
α-SiAlON	102	0.1596	0.2160	
	210	0.1418	0.1680	
β-SiAlON	101	0.1608	0.2160	
-	210	0.1274	0.1920	

Fig. 1 shows lower magnification electron micrographs of the irradiated α -SiAION specimens. No microcracks or voids/bubbles were observed along the grain boundaries in both α - and β -SiAION. HREM micrographs of the irradiated β -SiAION observed along the [001] direction without an objective aperture are presented in Fig. 2. Hexagonal atomic configurations of structures were observed. Clearly, all of atomic layers were straight along the <010> and the <110> directions without any bend or curve. In addition, no clear distortion of periodicity was observed. The microstructure of the present study was different from those of highly neutron-irradated β -Si₃N₄ up to 2.8x10²⁶ n/m² [4,5,6]. The presence of dislocation loops inside grains of both SiAIONs cannot be confirmed from these micrographs.

Although the irradiated specimens expanded, both dislocation loops and voids/bubbles were not observed. It can be suggested that the irradiation fluence up to 8.5×10^{24} n/m² (E>0.1 MeV) at 563 K induced only point defects or small clusters in SiAlON and Si₃N₄ ceramics, because of its lower fluence and lower temperature. Therefore, main reason for swelling should be attributed for the presence of point defects or small clusters of them in the irradiated specimens. The reasons for larger anisotropy in the lattice



Fig. 1 TEM micrograph of $\alpha\mbox{-SiAlON}$ irradiated to $8.5 x 10^{24} \mbox{ n/m}^2$ (E>0.1

MeV) at 563 K.



Fig. 2 HREM micrograph and electron diffraction pattern of irradiated β -SiAlON specimens observed along the [001] direction.

parameters of SiAlONs are discussed later. At present, the ason for small mismatch between the unit cell volume change and the macroscopic volume change of these ceramics is not clear. However, it may be contributed to internal strain caused by the anisotropic change in the lattice parameters [7].

Considering the macroscopic volume change and the unit cell volume change of two silicon nitride ceramics, α phase expanded about twice as much as β -phase. This can be attributed to the difference in the crystal structures of α and β -silicon nitride. The unit cell of β -Si₃N₄ consists of Si₆N₈, six-membered rings of SiN₄ tetrahedron. The crystal structure of α -Si₃N₄ was determined to be closely related to that of β -Si₃N₄, consisting of alternate stacking of the basal layers of β -Si₃N₄, accounting for the double c-axis dimension and a unit cell composition is $Si_{12}N_{16}$. Comparing β - and α -silicon nitride structures, the large interstices surrounded by the six-membered rings are open and continuous along the c-axis in β -silicon nitride while the interstices are closed (capped) in α -silicon nitride. If the knocked-on atoms positioned in capped interstices, the distortion of surrounding lattice should be larger than the case of the open channels. Therefore, expansion of α silicon nitride may be larger than that of β -silicon nitride. The change in lattice parameters of silicon nitride probably depends on the microstructural change, i.e., fluence. If interstitial dislocation loops were formed densely in particular planes, the change in lattice parameters should be modified, as reported in heavily irradiated β-silicon nitride, and the a-axis expanded and the c-axis contracted [3].

The lattice parameter change of both α - and β -SiAlON showed a complicated trend, i.e., the a-axis slightly expanded while the c-axis slightly shrank. The values of the a-axis expansion and the c-axis contraction were bigger in β -SiAlON. It is similar to the results obtained in our previous study for β -Si₃N₄ of 2.8-4.2x10²⁶ n/m² at 753 or 1003 K, i.e., 0.02~0.06% expansion in the a-axis and

0.03~0.06% contraction in the c-axis [3]. However, Hurley and Cocks [7] reported almost no change in Si₃N₄ while the a-axis shrinkage (-0.17%) and the c-axis expansion (+0.16%) for SiAlON after irradiation up to $3x10^{25}$ n/m² (E>0.1 MeV) at 1015 K, which were different from our results. SiAlONs contain larger ions such as Al and O than Si and N. Moreover, α -SiAlON contains extra-large cations in capped interstices in the structure. Then lattice expansion of SiAlONs induced by Frenkel pairs should be larger than those of silicon nitrides. The expansion of the a-axis and contraction of the c-axis in both SiAlONs may be caused by anisotropic structural relaxation derived from the presence of Frenkel defects, particularly interstitials located at the open or capped channels.

4. Conclusions

Neutron-irradiated Si₃N₄ and SiAlON ceramics, both α - and β -phases, were examined after concurrent irradiation up to 8.5×10^{24} n/m² (E>0.1 MeV) at 563 K. Changes in their macroscopic lengths and lattice parameters by the neutron irradiation were investigated along with their microstructural changes. The conclusions are as follows:

1. Changes in macroscopic length expansion of α - and β -SiAlON ceramics and α -Si₃N₄ were similar and almost double that of β -Si₃N₄.

2. Lattice parameter increased on both axes for both α and β -Si₃N₄. On the other hand, for SiAION ceramics, the a-axis increased while the c-axis shrank in both phases. However, the unit cell volume of both SiAIONs expanded. 3. The macroscopic volume changes calculated from the macroscopic length changes were slightly larger than the unit cell volume changes calculated from the lattice parameter changes in four examined specimens.

4. No voids, dislocation loops or grain boundary microcracks were observed from HREM observation in both neutron irradiated-SiAlONs.

[1] A. Rueanngoen, K. Kanazawa M. Akiyoshi, M. Imai, K. Yoshida and T. Yano, "Effects of neutron irradiation on polymorphs of silicon nitride and SiAlON ceramics", J. Nucl. Mater., 442, S394-S398 (2013)

[2] M. Akiyoshi, T. Yano, "Neutron-irradiation effect in ceramics evaluated from macroscopic property changes in as-irradiated and annealed specimens", Prog. Nucl. Ener., 50 (2008) 567-574

[3] T. Yano, M. Akiyoshi, K. Ichikawa, Y.Tachi and T. Iseki, "Physical Property Change of Heavily-Neutron-Irradiated Si_3N_4 and SiC by Thermal Annealing", J. Nucl. Mater., 289, 102-109 (2001).

[5] M. Akiyoshi, T. Yano and M. L. Jenkins, "A New Type of Defect on $\{11\ \overline{2}0\}$ Planes in β -Si₃N₄ Produced by Neutron Irradiation", Philos. Mag. Lett., 81[4], 251-258 (2001).

[6] M. Akiyoshi, K. Ichikawa, T. Donomae and T. Yano, "Macroscopic Properties and Microstructure Changes of Heavily Neutron-Irradiated β -Si₃N₄ by Annealing", J. Nucl. Mater., 307-311, 1305-1309 (2002).

[7] G.F. Hurley, F.H. Cocks, "X-ray analysis of internal strain in neutron-irradiated silicon nitride and oxynitrides", Am. Ceram. Soc. Bull. 60, 1302-1305 (1981).

References

^[4] M. Akiyoshi, T. Yano and M. L. Jenkins, "A Structural Model of Defects in β -Si₃N₄ Produced by Neutron-Irradiation", Philos. Mag. A, 81[3], 683-697 (2001).

B.9

Improvement of Thermal Conductivity of Silicon Nitride Ceramics by Heat-Treatment

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1. Introduction

Silicon nitride (Si_3N_4) has been known as known as a material with superior mechanical and thermal properties, and it has been expected to be used for high-temperature structural applications. In nuclear fields, Si₃N₄ ceramics have been considered as a candidate for confinement of minor actinides as an inert matrix [1]. Basically, it is so difficult to densify Si₃N₄ ceramics without high pressure or sintering additives because of the covalent nature of the Si-N bond. Therefore, some oxides are added to Si₃N₄ as sintering additives for densification of Si₃N₄. In general, densification of Si₃N₄ with these oxide additives proceeds via liquid phase sintering, i.e. the oxides react with Si₃N₄ and SiO_2 on the surface of Si_3N_4 at high temperature, and then liquid phase forms. Whereas some of the components from sintering additives dissolve into Si₃N₄ grains during sintering, most of the components from sintering additives remain at grain boundaries after sintering. Thermal and mechanical properties of Si₃N₄ are greatly related with grain boundary phases.

It has been reported that ideal thermal conductivity of single-crystal α - and β -Si₃N₄, along the *a*- and *c*-directions, is approximately 105 and 225, and 170 and 450 W/m•K at 27°C, respectively. However, polycrystalline Si₃N₄ ceramics generally show the thermal conductivity less than 100 W/m•K since their microstructure (e.g. grain boundary, pore and grain size) and crystalline imperfection (point defect, solid solution, stacking faults and dislocation) affect their thermal conductivity. In our previous study, Si₃N₄ was pressureless-sintered using MgO, Y₂O₃ and SiO₂ as sintering additives, and grain boundary phases were removed from Si_3N_4 by heat-treatment in order to investigate the effect of grain boundary phase on the thermal conductivity of Si₃N₄ ceramics [2]. It was found that thermal conductivity of Si₃N₄ was improved from 37 to 90 W/m•K by decreasing the amount of grain boundary phases after heat-treatment. However, not only the reduction of grain boundary phase from Si₃N₄ but also large grain growth of Si₃N₄ occurred during heat-treatment. It has been experimentally demonstrated that thermal conductivity of polycrystalline ceramics increases with an average grain size [3]. For this reason, it was not clear whether the reduction of grain boundary phase by heat-treatment would be effective for the improvement of the thermal conductivity of Si_3N_4 ceramics or not.

In this study, α - or β -Si₃N₄ powder with larger grain size was uses as a starting material, and the effect of heat-treatment on thermal conductivity of Si₃N₄ ceramics

using MgO, Y_2O_3 and SiO₂ as sintering additives was investigated in terms of their microstructure and the amount of grain boundary phase.

2. Experimental Procedures

Alpha-Si₃N₄ powder (SN-7, average grain size : 4.3 μm, $\alpha/(\alpha + \beta)$ phase ratio : 73%, Denki Kagaku Kogyo, Japan) and β -Si₃N₄ powder (SN-F1, average grain size : 2.4 μ m, $\beta/(\alpha + \beta)$ phase ratio : 95%, Denki Kagaku Kogyo, Japan) were used as starting materials, and 3 wt% of SiO₂ (SEAHOSTAR KE-P30, average grain size : 0.25-0.31 µm, Nippon Shokubai, Japan), 3 wt% of MgO (MJ-30, average grain size : 0.35 µm, Iwatani Kagaku Kogyo, Japan) and 4 wt% of Y₂O₃ (RU, average grain size : 1.2 µm, Shin-Etsu Chemical, Japan) were added to Si₃N₄ powder as sintering additives. The powder was mixed by wet ball-milling, and then dried using a rotary evaporator. The powder was crushed using a mortar, followed by sieving through the opening of 200 µm. The powder was formed into green compacts by uniaxial pressing of 20 MPa, and then cold isostatically pressed at 200 MPa. The green compacts were embedded into bed powder consisting of α -Si₃N₄ and boron nitride powder (the weight ratio of α -Si₃N₄/BN was 1.0) and pressureless-sintered at 1650°C for 1-4 h in nitrogen flow (pressureless-sintered Si₃N₄ was denoted as "sintered Si_3N_4 " in this report). The sintered Si_3N_4 ceramics were heat-treated at 1950°C for 8 h under a nitrogen pressure of 1.0 MPa (sintered Si₃N₄ after heat-treatment was denoted as "heat-treated Si3N4" in this report).

Bulk density (ρ) of sintered Si₃N₄ and heat-treated Si₃N₄ was measured by Archimedes' method. Crystalline phases of sintered Si₃N₄ and heat-treated Si₃N₄ were indentified by X-ray diffractometry (XRD). Microstructure of sintered Si₃N₄ and heat-treated Si₃N₄ after plasma etching was observed by scanning electron microscope (SEM). Oxygen contents in Si₃N₄ ceramics were determined with a nitrogen/oxygen determinator. Thermal diffusivity (α) and specific heat (C_p) were measured by laser flash method, and thermal conductivity (κ) was calculated from the following equation:

 $\kappa = \alpha \bullet C_p \bullet \rho \tag{1}$

3. Results and Discussion

Figure 1 shows the change in bulk density of sintered Si_3N_4 with sintering time. As Si_3N_4 was pressureless-sintered at 1650°C for sintering time more

than 2 h, their bulk density was almost constant, and the



Fig. 1 The change in bulk density of sintered Si_3N_4 using SN-7 (open square) and SN-F1 (open circle) powder with sintering time. Bulk density of Si_3N_4 at zero in sintering time corresponds to green density of Si_3N_4 before sintering.



Fig. 2 The mass loss and oxygen content of sintered Si_3N_4 and heat-treated Si_3N_4 using SN-7 (open square) and SN-F1 (open circle) powder.

value of Si_3N_4 ceramics pressureless-sintered for 2 h was 2.91 g/cm³ and 3.04 g/cm³, respectively. In this study, Si_3N_4 ceramics pressureless-sintered at 1650°C for 2 h were selected and the sintered Si_3N_4 ceramics were heat-treated at 1950°C for 8 h under a nitrogen pressure of 1.0 MPa. After heat-treatment, bulk density of sintered Si_3N_4 increased to 3.13 g/cm³.

Figure 2 shows the mass loss and oxygen content of sintered- and heat-treated Si_3N_4 . In sintered Si_3N_4 , mass loss linearly increased with an increase in sintering time, and thus oxygen contents decreased. After heat-treatment of Si_3N_4 ceramics sintered at 1650°C for 2 h, mass loss increased to approximately 16 %, and oxygen content of Si_3N_4 ceramics using SN-7 and SN-F1 powder significantly decreased from 3.5 and 3.3 % to 0.64 and 0.71 %, respectively.

SEM micrographs of microstructure of sintered- and heat-treated Si_3N_4 are exhibited in Fig. 3. Sintered Si_3N_4 using SN-7 powder consisted of relatively fine grains, and its grain size was much smaller than that of sintered Si_3N_4 using SN-F1. After heat-treatment at 1950°C, large grain growth occurred in Si_3N_4 ceramics using SN-7 and grain size of heat-treated Si_3N_4 became much larger than that of sintered Si_3N_4 . On the other hand, grain growth was not observed in heat-treated Si_3N_4 ceramics using SN-F1 powder, i.e. grain size of heat-treated Si_3N_4 was nearly the same as that of sintered Si_3N_4 . In both Si_3N_4 ceramics using SN-7 and SN-F1 powder, it seemed that the amount of grain boundary phase would be significantly reduced after heat-treatment.

XRD patterns indicated that sintered Si₃N₄ using SN-7 and SN-F1 mainly consisted of β -Si₃N₄ but it contained small amount of α -Si₃N₄. Diffraction peaks derived from sintering additives were not observed in XRD patterns of sintered Si₃N₄. In the case of heat-treated Si₃N₄, small amount of α -Si₃N₄ existed in sintered Si₃N₄ was transformed into β -Si₃N₄, and it mainly consisted of β -Si₃N₄. In addition, diffraction peaks corresponding to some crystalline phases (Y₂O₃ and Y₂Si₃N₄O₃) derived from sintering additives were observed in XRD patterns of heat-treated Si₃N₄.

The results of SEM observation, oxygen contents and XRD suggested that most of the components derived from sintering additives existed as glassy phase in sintered Si_3N_4 , and the amount of glassy phase significantly decreased after heat-treatment at 1950°C. The small amount of glassy phase existed in Si_3N_4 ceramics was crystallized as Y_2O_3 and $Y_2Si_3N_4O_3$ after heat-treatment.

Figure 4 shows the relationship between thermal conductivity and oxygen content of sintered- and heat-treated Si₃N₄. Thermal conductivity of sintered Si₃N₄ linearly increased with an increase in sintering time, i.e. the increase in their thermal conductivity resulted from the decrease in oxygen content, and the values of sintered Si₃N₄ using SN-7 and SN-F1 powder were in the range 30.0 to 48.3 W/m•K and 24.2 to 42.0 W/m•K, respectively. Si₃N₄ ceramics pressureless-sintered at 1650°C for 2 h were heat-treated at 1950°C for 8 h, and their thermal conductivity was measured. In the case of Si₃N₄ ceramics using SN-7 powder, thermal conductivity of heat-treated Si_3N_4 was around twice of the value of sintered Si_3N_4 , and the thermal conductivity was increased from 41.4 to 87.2 W/m•K. In this case, large grain growth occurred during heat-treatment, and the grain size of heat-treated Si₃N₄ became so large compared with sintered Si₃N₄, and the improvement of the thermal conductivity of Si₃N₄ would be attributed to not only the reduction of grain boundary phase but also the grain growth. In the case of Si₃N₄ using SN-F1 powder, thermal conductivity of Si₃N₄ ceramics was also significantly increased from 36.0 to 73.2 W/m•K after heat-treatment. In this case, the reduction of grain boundary phase mainly affected the thermal conductivity of Si₃N₄ ceramics because the grain size of heat-treated Si_3N_4 was nearly the same as that of sintered Si_3N_4 . Furthermore, bulk density of both Si₃N₄ ceramics using SN-7 and SN-F1 powder slightly increased after heat-treatment, and this would have small effect on the improvement of thermal conductivity. Heat-treated Si₃N₄ using SN-7 powder showed higher thermal conductivity than heat-treated Si₃N₄ using SN-F1 powder, and the difference in thermal conductivity between heat-treated Si_3N_4 using SN-7 and



Fig. 3 SEM micrographs of microstructure of sintered Si_3N_4 (a, c) and heat-treated Si_3N_4 (b, d) using SN-7 (a, b) and SN-F1 (c, d) powder.



Fig. 4 The relationship between thermal conductivity and oxygen content of sintered Si_3N_4 and heat-treated Si_3N_4 using SN-7 (open square) and SN-F1 (open circle) powder.

SN-F1 powder might result from the difference in starting powder (α -Si₃N₄ and β -Si₃N₄). However, the reason for this result has not been clear yet. From the results, it was concluded that the reduction of grain boundary phase from Si₃N₄ was effective for the improvement of their thermal conductivity in addition to grain growth of Si₃N₄.

4. Conclusion

In this study, α - or β -Si₃N₄ powder with larger grain size was uses as starting material, and the effect of heat-treatment on thermal conductivity of Si₃N₄ ceramics using MgO, Y₂O₃ and SiO₂ as sintering additives was investigated in terms of their microstructure and the amount of grain boundary phase. From the results of SEM observation, oxygen contents and XRD, it was suggested that most of the components derived from sintering

additives existed as glassy phase in sintered Si₃N₄, and the amount of glassy phase significantly decreased, and then small amount of glassy phase existed in Si₃N₄ ceramics crystallized as Y_2O_3 and $Y_2Si_3N_4O_3$ after was heat-treatment at 1950°C for 8 h under a nitrogen pressure of 1.0 MPa. Thermal conductivity of sintered Si₃N₄ linearly increased with an increase in sintering time, i.e. the increase in their thermal conductivity resulted from the decrease in oxygen content. In the case of Si₃N₄ ceramics using SN-7 powder, thermal conductivity of heat-treated Si₃N₄ was around twice of the value of sintered Si₃N₄, and the thermal conductivity was increased from 41.4 to 87.2 W/m•K. In this case, the improvement of the thermal conductivity of Si₃N₄ would be attributed to not only the reduction of grain boundary phase but also the grain growth. In the case of Si₃N₄ using SN-F1 powder, thermal conductivity of Si₃N₄ ceramics was also significantly increased from 36.0 to 73.2 W/m•K after heat-treatment. In this case, the reduction of grain boundary phase mainly affected the thermal conductivity of Si₃N₄ ceramics because the grain size of heat-treated Si₃N₄ was nearly the same as that of sintered Si₃N₄. From the results, it was concluded that the reduction of grain boundary phase from Si₃N₄ was effective for the improvement of their thermal conductivity in addition to grain growth of Si₃N₄.

References

[1] T. Yano, J. Yamane, K. Yoshida, S. Miwa, M. Ohsaka: Mater. Sci. Eng., Vol.9, 012024 (2010).

[2] Y. Sekimoto, T. Wasanapiarnpong, M. Imai, K. Katayama, T. Yano: Key Eng. Mater., Vol.352, pp. 35-39 (2007).

[3] K. Yoshida, M. Imai, T. Yano : High Temperature Ceramic Matrix Composites, edited by W. Krenkel, R. Naslain, H. Schneider, Wiley-VCH, pp. 388-393 (2001).

B.10 Creation of Thermoresponsive Polymer-Clay Hybrid Nanosheets

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A certain class of layered inorganic materials has alternative laminated structures consisting of inorganic monocrystalline layers and exchangeable metal ions, the former of which can be modified with organic molecules *via* a grafting reaction. Inorganic monocrystalline layers in layered inorganic materials can be utilized as nanosheets upon exfoliation, and nanosheets modified with organic molecules are attracting attention because of their possible use as organically compatible nanofillers, catalytic nanofilms, and biosensing nanomaterials.

One attractive possible strategy toward dynamic control of nanosheet surface properties is surface modification of nanosheets with stimuli-responsive polymers, which have latently changeable properties in response to external stimuli. Such nanosheets would be useful nanomaterials for on-demand systems with analytical and biomedical applications such as separation matrices and drug delivery carriers. In particular, thermoresponsive poly(N-isoproylacrylamide) (PNIPAAm) is recognized as an appropriate functional polymer. We have already synthesized PNIPAAm-based hybrid polymer materials, which make it possible to separate mutually actinides and lanthanides [1,2]. Herein, in order to enhance the separation functions of the PNIPAAm-based polymer, we tried to create novel polymer-clay hybrid nanosheet. As shown in Figure 1, the hybrid nanosheet consisting of PNIPAAm polymer on the interlayer surfaces of a Dion-Jacobson-type layered peroviskite HLaNb₂O₇·xH₂O (HLaNb) was synthesized by surface-initiated atom transfer radical polymerization (SI-ATRP).



Figure 1. Schematic illustration of synthesis processes of PNIPAAm-HLaNb nanosheet.

Firstly, ATRP initiator, Phosphonomethyl m-(chloromethyl)phenylmethylate (PMCP), was immobilized onto the *n*-decoxy derivative of HLaNb. And then, the PMCP-modified HLaNb was added into the solution, in which CuCl, CuCl₂, and tris[2-(dimethylamino)ethyl]amine (Me₆TREN) were

dissolved in degassed 2-propanol containing NIPAAm (10 mol/L) and vinylpyrrolidone (2 mol%) monomers. The SI-ATRP reaction could be proceeded at room temperature for 24 h, and quenched by exposing the solution to air. The crude product was then rinsed repeatedly with methanol, and dried under a vacuum after centrifugation.

In order to confirm the presence of PMCP and PNIPAAm onto HLaNb nanosheets, X-ray diffraction (XRD) patterns were measured using CuKa and/or FeK α radiations. The results are shown in Figure 2. In the XRD pattern of PMCP-HLaNb, the diffraction line corresponding to their interlayer distance (d = 2.85 nm) was changed in comparison with that of *n*-decoxy derivative of HLaNb (d = 2.73). The (100) and (110) reflections of HLaNb do not shift in either pattern, indicating that the structure of the perovskite-like slabs is retained after the reaction. On the other hand, the (100)and (110) reflections of HLaNb in PNIPAAm-HLaNb were present, while no diffraction lines at low angles were observed. This fact indicates that PNIPAAm polymer chains covered the interlayer surfaces of HLaNb.



Figure 2. XRD patterns of (a) C10-HLaNb, (b) PMCP-HLaNb, and (c) PNIPAAm- HLaNb.

The PNIPAAm-HLaNb configurations were measured using transmission electron microscopic (TEM) and atomic force microscopic (AFM) images. Plate-like morphology with light contrast was observed from the TEM image, and the electron diffraction pattern could be indexed as a pattern along the [010] zone axis for an orthorhombic cell (a = 0.39 nm). The results suggest that the PNIPAAm-HLaNb was exfoliated into nanosheets. Moreover, AFM images showed that almost all the PNIPAAm-HLaNb exhibited

plate-like morphology with a high aspect ratio, and its thickness was ca. 25 nm as shown in **Figure 3**.

To clarify the phase transition behavior of PNIPAAm-HLaNb nanosheets, the transmittance of the aqueous suspension dispersed with the nanosheets (1 mg/mL) at 400 nm was continuously monitored at a heating and cooling rates of 0.5°C/min using a UV-visible spectrometer (Figure 4). Apparent cloud point of PNIPAAm-HLaNb nanosheet can be determined by absorbance change at 400 nm. A significant increase in absorbance of the aqueous suspension of PNIPAAm-HLaNb nanosheet was observed at temperatures higher than 34°C, indicating that the nanosheets were aggregated in water due to the coil-to-globule transition of the modified PNIPAAm chains. The phase transition behavior and transition temperature were consistent with those of a PNIPAAm aqueous solution, indicating that preparation of thermoresponsive nanosheets can be achieved by surface modification of HLaNb with NIPAAm through SI-ATRP. Around the phase transition temperature of 34°C, hysteresis of the changes in absorbance was observed between heating and cooling cycles. Although the nanosheets are loosely aggregated with increases in temperature by their hydrophobic interaction, dispersion of the nanosheets during cooling process is dominated by their self-diffusion, leading to the hysteresis. At 25 and 50°C, no notable differences in absorbance are observed between heating and cooling cycles, indicating that the nanosheets exhibited excellent reversibility during phase transition. Interestingly, the nanosheet suspension at high temperatures maintains a cloudy state, even with increases in temperature.

In previous studies on PNIPAAm-modified nanoparticles with 200 - 300 nm diameter, precipitates appeared at high temperatures by their aggregations *via* the hydrophobic interaction. In contrast, aqueous dispersion of PNIPAAm-modified nanoparticles with 70 nm in lateral dimensions exhibited continuous low transmittance above the phase transition temperature. In this study, PNIPAAm-HLaNb nanosheets had sub-micron sizes in lateral dimensions with a thickness of ca. 25 nm, as shown in Figure 3. Since the precipitation of PNIPAAm-HLaNb nanosheets was not observed with an increase in temperature, the thickness of nanosheets would influence on the stability of the aqueous dispersion at high temperature.

In summary, PNIPAAm-HLaNb nanosheets were prepared by exfoliation from their layer structure through SI-ATRP. The nanosheets exhibited a plate-like morphology with a high aspect ratio in which their lateral size is mostly sub-micron ordered and their thickness is ca. 25 nm. The phase transition behavior of the PNIPAAm-HLaNb nanosheets dispersed in water can be observed with increases in temperature to above 34 °C. The cloudy state of the nanosheet suspension at high temperatures was relatively stable in spite of the hydrophobic PNIPAAm chains modified on the surfaces. These results suggest that surface modification with stimuli-responsive polymers is a possible technique for preparation of functional nanosheet materials.



Figure 3. AFM image and the cross-sectional profile of PNIPAAm-HLaNb nanosheet.



Figure 4. Temperature dependence in absorbance at 400 nm in aqueous suspension of PNIPAAm-HLaNb nanosheets. Symbol; open circles: heating cycle, closed circles: cooling cycle.

References

[1] T. Tsukahara, et al., *React.Funct.Poly.*, 79, 36 (2014).

[2] T. Tsukahara, et al., Chem. Lett., 1381 (2011).

B.11 Uranyl-halide Complexation in *N*,*N*-Dimethylformamide: Halide Coordination Trend Manifests Hardness of [UO₂]²⁺

Koichiro TAKAO

1. Introduction

Pearson's hard soft acids bases (HSAB) principle is very simple, but quite helpful in describing interactions of metal ions with ligands. Although a uranyl ion $([UO_2]^{2+})$, the most relevant species in nuclear engineering chemistry, is frequently regarded as a hard acid, its coordination strength with halides (Cl⁻, Br⁻, I⁻, here symbolized with X⁻) in different media (e.g., aqueous solutions and ionic liquids) does not clearly follow the sequence of hardness of these ligands, except for F⁻. In this context, use of an appropriately selected organic solvent may help to more clearly differentiate the coordination strength of $[UO_2]^{2+}$ -X⁻ complexes. Here, N,N-dimethylformamide (DMF) has been employed as a solvent, because use of the isolable $[UO_2(DMF)_5](ClO_4)_2$ as a source of $[UO_2]^{2+}$ will most simplify the system.

2. Experimental

2.1.Sample Preparation

All the chemicals used in this study were of reagent grade. Pentakis(N,N-dimethylformamide)dioxouranium perchlorate, $[UO_2(DMF)_5](CIO_4)_2$, was prepared from a reaction between $UO_2(ClO_4)_2 \cdot 5H_2O$ and DMF in triethyl orthoformate, followed by recrystallization from CH₂Cl₂/ether. Tetra-*n*-butylammonium ([TBA]⁺) salts of Cl⁻, Br⁻, and I⁻ were recrystallized from ethanol/ether prior to use. The stock solutions of $[UO_2(DMF)_5](CIO_4)_2$ and [TBA]X of each X⁻ system were indivisdually prepared in DMF for the UV-vis experiments. As EXAFS samples, aliquots of DMF solution dissolving [UO₂(DMF)₅](ClO₄)₂ (40 mM) and different amount of [TBA]X were confined in disposable plastic cuvettes with silicone rubber septa. 2.2 Methods

UV-vis absorption spectra of $[UO_2]^{2+}$ at different $[X^-]$ in DMF were recorded using J&M TIDAS 100 or Agilent 8453 photodiode array spectrophotometers. All measurements were performed at 295 ± 1 K. The obtained data were analyzed using the nonlinear least-squares regression program *HypSpec*.

X-ray absorption fine structure (XAFS) spectroscopy was performed at the Rossendorf Beamline (ROBL) BM20 at the European Synchrotron Radiation Facility (ESRF, Grenoble, France). Uranium L_{III} -edge X-ray absorption spectra of the DMF solutions containing $[UO_2]^{2+}$ and [TBA]X were recorded in transmission mode using argon-filled ionization chambers at 295 ± 1 K. The XAFS spectra of each sample was recorded twice or four times and merged. EXAFS data extraction and fits were processed with IFEFFIT. The threshold energy E_0 of the U L_{III} -edge was defined at 17185 eV.

3. Results and Discussion

3.1.UV-vis Titration

The UV-vis absorption spectra of the DMF solution dissolving [UO₂(DMF)₅](ClO₄)₂ (6.65 mM) at the different total concentrations of $Cl^{-}([Cl^{-}]_{tot})$ and 295 K are shown in Fig. 1. The characteristic fine structure arising from the ligand-to-metal charge transfer in $\left[\text{UO}_2\right]^{2+}$ was observed in this wavelength range. With increasing $[Cl]_{tot}$, the absorption intensity decreases, and the more finely structured spectrum occurs at the last points of titration. This series of the absorption spectra does not have any isosbestic points, implying that it cannot be explained by a single equilibrium reaction. The spectra at $[CI]_{tot} \ge 26.6$ mM are almost identical to that of $[UO_2Cl_4]^{2-}$. Thus, four stepwise complication reactions to form $[UO_2Cl_n]^{2-n}$ (n = 1-4) resulted in Fig. 1. Stability constants of the occurring species were derived from a non-linear least squares regression on the HypSpec program. The obtained logarithmic gross stability constants of $[UO_2Cl_n]^{2-n}$ (log β_n) at 295 K are log $\beta_1 = 9.67$, log $\beta_2 = 15.49$, log $\beta_3 =$ 19.89, and $\log \beta_4 = 24.63$.



Fig. 1 UV-vis absorption spectra of the DMF solution dissolving $[UO_2(DMF)_5](ClO_4)_2$ (6.65 mM) at different $[Cl^-]_{tot}$ and 295 K.

Using these quantities, the species distribution of each $[UO_2Cl_n]^{2-n}$ was calculated at different conditions as shown in Fig. 2.



Fig. 2 Species distribution diagram of $[UO_2Cl_n]^{2-n}$ (*n* = 0-4) in DMF solution ([[UO_2]²⁺]_{tot} = 6.65 mM) at 295 K.

In a similar manner, the coordination of Br⁻ and Γ to $[UO_2]^{2^+}$ was also examined. Although the coordination of Br⁻ to $[UO_2]^{2^+}$ was actually observed, the maximum number of *n* in $[UO_2Br_n]^{2^{-n}}$ was 2 under the studied condition $([[UO_2]^{2^+}]_{tot} = 6.07 \text{ mM}, [Br⁻]_{tot} = 0.28.8 \text{ mM}, 295 \text{ K})$. The estimated log β_n of $[UO_2Br_n]^{2^{-n}}$ for n = 1 and 2 are 3.45 and 5.42, respectively, which are much smaller than the corresponding values of $[UO_2Cl_n]^{2^{-n}}$. In contrast, no coordination of Γ was detected. In conclusion, the difference in the coordination strengths of X^- to $[UO_2]^{2^+}$ was clearly manifested in the DMF solvent systems.

3.2.EXAFS

The next concern in this study is the details about molecular structures of the occurring $[UO_2]^{2^+}$ -X⁻ complexes. The extended XAFS (EXAFS) spectra at different [Cl⁻]_{tot} afforded the radial structure functions shown in Fig. 3.



Fig. 3 Fourier transforms of k^3 -weighted U L_{III}-edge EXAFS spectra of the DMF solutions dissolving $[UO_2(DMF)_5](ClO_4)_2$ (40 mM) and [TBA]Cl (0-200 mM) at 295 K.

Using the gross stability constants of $[UO_2Cl_n]^{2-n}$ (n = 1-4) and the *HypSpec* program, the EXAFS spectra of the individual species were extracted from the titration series of the parent EXAFS spectra of Fig. 3. Finally, the detailed stepwise formation of $[UO_2Cl_n]^{2-n}$ (n = 0-4) was concluded as shown in Fig. 4.



Fig. 3 Complexation equilibria of $[UO_2]^{2+}$ with Cl⁻ in DMF. L denotes a DMF molecule. The bond distances (Å) found in EXAFS are also attached.

The same experiments and analyses were also performed for the $[UO_2]^{2+}$ -Br⁻ system in DMF. The resulting complexation scheme of $[UO_2Br_n]^{2-n}$ (n = 0-2) was summarized as displayed in Fig. 4.



Fig. 4 Complexation equilibria of $[UO_2]^{2+}$ with Br⁻ in DMF. L denotes a DMF molecule. The bond distances (Å) found in EXAFS are also attached.

4. Conclusion

In this study, the complexation of $[UO_2]^{2^+}$, which could be hard as a Lewis acid, with the X⁻ congeners like Cl⁻, Br⁻, and I⁻ in DMF was investigated by means of UV-vis absorption spectroscopy and EXAFS. As a result, the stability of $[UO_2]^{2^+}$ -X⁻ complexes in DMF is exactly in line with the hardness order of these X⁻ as expected. This trend clearly demonstrates the hardness of $[UO_2]^{2^+}$.

Reference

K. Takao, S. Takao, Y. Ikeda, G. Bernhard, C. Hennig; Dalton Transactions, Vol.42, pp. 13101-13111 (2013).

C. System and Safety Engineering

C.1 **Progress in Innovative Nuclear Reactor and Criticality Safety Study**

Toru OBARA

Several Studies on innovative nuclear reactors and criticality safety have been performed. It is intending to give concepts of nuclear reactors for various demands and needs in future and analyze the phenomena in criticality accident of fuel solutions.

1. Design study of small reactor for semiconductor production

A small reactor for NTD-Si was proposed in the study. It is intended to irradiate 12 inches silicon ingots. In the study, short length PWR fuel assemblies were used in the core. It was shown the reactor can achieve enough production rate of silicon semiconductors by using short length PWR fuel assemblies [1].

2. Study on thermal neutron filter for NTD-Si irradiation

To improve the performance in NDT-Si irradiation by research reactors, the concept of thermal filters using B_4C particles were proposed. A design of new irradiation holder was proposed and the performance was analyzed [2].

3. Study on particle type burnable poisons

In long-life prismatic HTGR, it is important to compensate excess reactivity effectively. The performance of particle type burnable poisons were analyzed. It was shown that the combination of two types of burnable poison can have excellent feature to make the excess reactivity small and flat in prismatic HTGR [3].

4. Study on passive decay heat removal by conduction and radiation in HTGR

The analysis was performed to show the possibility to remove decay heat from simplified pebble bed HTGR core by radiation and conduction. The reactor was set under the ground and the final heat sink of the decay heat was soil in the underground. It was shown the proposed concept of small simplified pebble bed HTGR has a capability to remove decay heat after the shutdown by the radiation and conduction only [4].

5. Design study of small long-life HTGR free from prompt supercritical accidents

A design concept of small long-life HTGR core was proposed. It utilizes particle type burnable poisons to suppress excess reactivity. In addition to that, to avoid any prompt critical accident, the reactor core temperature is controlled properly. The decrease of thermal efficiency was estimated by the decrease of core temperature and it was shown the decrease was acceptable. In the reactor concept, it is possible to be free from prompt criticality accident during operation [5].

6. Prismatic HTGR core free from graphite oxidation in air ingress accidents

In the study, SiC coated fuel element block is employed in prismatic HTGR to prevent graphite oxidation in air ingress accident. The impact of the coating was estimated by neutronic and thermal hydraulic analysis. It was shown the impact was small in burnup performance and heat removal in operation [6].

7. Effect of neutron coupling between the fuel solution tanks in criticality accident

Numerical analysis method based on the integral kinetic model was applied to the kinetic analysis in criticality accidents in two fuel solution tanks system. The neutron coupling effect between the tanks can be treated by the model. The reactivity feedback effect by radiolytic gas void was included in the analysis. It was shown the increase of integral power in the accident could be estimated based on the results of analysis [7].

8. Development Monte Carlo based code for burnup analysis in OTTO cycle pebble bed reactors

A new code was developed to analyze burunup performance of Once-Through-Then-Out (OTTO) cycle pebble bed reactor. The code is based on Monte Carlo method in neutron transport calculation, so the exact treatment of the baps between the ball and the cavity at the core top [8].

REFERENCES

1. Byambajav Munkhbat, Toru Obara, Journal of Nuclear Science and Technology, 50 [1], pp.46-58 (2013).

2. Masao Komeda, Kozo Kawasaki, Toru Obara,

Applied Radiation and Isotopes, 74, 70-77, (2013)

3. Toru Obara, Taiki Onoe, Annlas of Nuclear Energy, 57, 216-220 (2013).

4. Dwi Irwanto, Toru Obara, Annlas of Nuclear Energy, 60, 383-395 (2013).

5. Piyatida Trinuruk, Toru Obara, Journal of Nuclear Science and Technology, 50 (9), 898-912 (2013).

6. Piyatida Trinuruk, Toru Obara, Annals of Nuclear Energy, 63, 437-445 (2013).

7. Toru Obara, Haruka Kikuchi, Transactions of the American Nuclear Society, 109, 888-889 (2013).

8. Topan Setiadipura, Toru Obara, Transactions of the American Nuclear Society, 109, 1431-1433 (2013).

C.2 Positional Stabilization of Torus Plasma with Simple Helical Coils

Shunji TSUJI-IIO, Hiroaki TSUTSUI

In tokamaks, elongated plasmas for good confinement suffer from so-called vertical displacement events (VDEs) during disruptions. On the other hands, it has been shown that the plasma position is robustly stable in a currentcarrying stellarator.¹⁾ In order to suppress VDEs, we proposed saddle shaped stellarator system, which is composed of four coils on the outside of a vacuum vessel. The objective of our study is to investigate the principle and to improve the configuration of the coils for vertical stability experimentally.

We designed and have been constructing a small tokamak device (R = 0.33 m, a = 0.09 m, $B_t = 0.3$ T) which has an elongated cross section to demonstrate above mentioned effect. Among the progress of the construction, we report the following topics: strength tests of a trial toroidal field coil and discharge tests of flywheel power supply for poloidal field coils.

Firstly, we designed support structures for the toroidal field coils by stress analysis using the finite element method. After we confirmed soundness of the support structure numerically, we performed strength tests on a trial TF coil as shown in Fig. 1. The maximum electromagnetic force which was estimated to be a centering force of 550 kgf was simulated by the load of a heavy weight of 1000 kg in the tests. It was found from the strain measurements that the deformation of the support structure can be elastic even under overload up to twice of the estimated maximum centering force. After we got the result, we made all sixteen toroidal field coils and assembled them.

We proposed a new simple flywheel generator system inspired by micro hydro power technology as the power supply for poloidal field coils (Fig. 2).²⁾ This system uses a self-excited induction generator (SEIG) converted from a commercially available induction motor so that it can be cost effective. The SEIG can maintain nearly constant terminal voltage in combination with rectifier circuit much longer than commonly used capacitor banks. The post stage dc-chopper which was controlled by taking voltage fluctuations in SEIG into account converts the varying voltage to arbitrary voltage to control currents flowing in poloidal field coils. We conducted discharge test of the power supply with a sample coil comparable to poloidal field coils (Fig. 3). It was demonstrated that the proposed power supply can maintain command squarewave current flowing in the test coil for 1 s which is long enough to our small tokamak experiments.

References

1) H. Ikezi et. al., Phys. Fluids 22, 2009 (1979).

2) S. Hatakeyama et. al., 1PoCL-07, MT-23, 2013.

strain gage

Fig. 1 Pictures of strength tests of a trial toroidal field coil.



Fig. 2 Circuit configuration of the flywheel generator system.



Fig. 3 Experimental waveforms of the sample coil current and the generator voltage.

C.3

Ultrasonic Velocity Profiler for Very Low Velocity Field

Hiroshige Kikura and Nobuyoshi Tsuzuki

1. Introduction

High Level radioactive Waste (HLW) will be reprocessed and disposed geologically in Japan. High Level radioactive Liquid Waste (HLLW) from HLW is mixed with molten glass in a glass melter, and the glass is poured into a canister, then the canister will be brought into a waste disposal site. However, the operation of the glass melter is not easy. Flow behavior of molten glass in the glass melter is very complicated and slow. The complexity means difficult to understand and the sloth means difficult to measure. Due to the opaqueness and high temperature of molten glass, Ultrasonic Velocity Profiler (UVP) method with buffer rod is considered as most applicable for the measurement of the glass melter. However, the slow velocity field of the molten glass is close or even below the lowest detectable velocity that the usual UVP method can detect. Consequently, a new algorithm and measurement system is required to understand the flow behavior in the glass melter.

2. Phase difference method

In the conventional UVP measurement, the velocity component along the measurement line is calculated from Doppler frequency. However, devices for the UVP measurement are digitized and based on digitized time domain, this corresponds to the minimum detectable velocity, namely a velocity threshold. The time resolution, $\Delta T_{\rm DS}$, can be expressed as

$$\Delta T_{\rm DS} = \frac{N_j}{f_{\rm PRF}} \,,$$

where N_j is the number of emitted ultrasonic pulses and f_{PRF} is pulse repetition frequency.

In fact, an ultrasound echo signal even from tracer particles can be expressed in a sinuous curve form, and it means 'phase' will be calculated for each echo signal. If there is the second emission, the echo signal is represented using a slightly different value of phase, which reflects the motion of tracer particles inside the measurement volume. The main idea of a new method is to detect the mean particle displacement, Δx , from the difference of the phase of two successive signals, $\Delta \theta$, as,

$$\Delta x = \frac{c}{2\pi f_0} \Delta \theta,$$

where c is sound velocity and f_0 is basic frequency of the emitted ultrasound.

Since this technique estimates velocity from only two successive repetitions of echo reception, the minimum time resolution of this new 'phase difference method,' ΔT_{PD} is given as,

$$\Delta T_{PD} = \frac{2}{f_{PRF}} \, .$$

The maximum velocity that can be detected, V_{max} , is the same as for the Doppler method because the range of $\Delta\theta$ remains between $-\pi$ and π . Numeric simulations suggest that the velocity threshold of the phase difference method can be affected by quantization error. Therefore, the practical performance of the phase difference method should be validated.

3. Experimental verification

The capability of measuring velocity profiles was investigated using the flow inside a rotating cylinder. Figure 1 shows a schematic of the experimental set up. The acrylic rotating cylinder has an outer diameter of 160 mm and a wall thickness of 3 mm. The cylinder is set up in a water tank with temperature controlled at 30°C, thus, sound speed in the cylinder is 1550 m/s. Tracer particles are dispersed in the fluid. An ultrasonic transducer with 4 MHz basic frequency and 5 mm element diameter is fixed at the position L = 2.5mm in Fig. 1. The fluid in the cylinder is in rigid body motion, thus, measured velocity component along measurement line should be constant on the line.

Figure 2 shows a color density plot of the evolution of the velocity profile after a start of cylinder rotation. The velocity field evolves from the near wall region and observed the velocity profile from transient state to a steady state agreeing with theoretical flow behavior. Consequently, the phase difference method can obtain a very low velocity field and the method showed applicability for velocity measurement of the glass melter.



Fig. 1. Top view of the experimental apparatus.



Fig. 2. Velocity profile obtained using phase difference method.

Reference

1. T. Ihara, et al.; Flow Measurement and Instrumentation, Vol. 34, pp. 127-133 (2013).

C.4 Beam Focusing for Air-coupled Ultrasonic Flowmeter

Hiroshige Kikura and Nobuyoshi Tsuzuki

1. Introduction

The flowrate measurement is applied in the controlling and monitoring industrial processing. In the nuclear power plants, the feed water flowrate is monitored to control the thermal output, and the flow is high pressure and high temperature. In such severe condition, flow measurement method is limited. The conventional measuring equipment is orifice flowmeter. Orifice flowmeter is simple and capable to apply in high temperature condition. However, the orifice flowmeter requires maintenance and re-calibration. For these concerns, ultrasonic flowmeter has been used recently. The ultrasonic flowmeter is capable to apply non-invasive measurement and on-line maintenance. Besides, the acoustic technique development enables higher measurement accuracy. However, the Piezo-electric element in the ultrasonic sensor has difficulty in applying high temperature condition of the thermal or nuclear power plants. In the higher temperature condition than Curie-temperature of the material, the ultrasonic sensor loses the piezoelectricity and causes the desensitization. On the other hand, 'air-coupled ultrasound' applies propagation in the air, and into the test bodies. In the method, sensors are not required of contact to the test bodies. For this reason, air-coupled ultrasonic flow metering is developed and verified about the applicability.

2. Air-coupled ultrasonic flowmeter



Fig. 1 Air-coupled ultrasonic flowmeter.

The air-coupled flowmeter is placed far from the wall of the pipe. Ultrasound is emitted from upstream sensor #1 (right side of Fig. 1), propagates through the air, pipe wall, working fluid (in this case, water), pipe wall again, the air, and the ultrasound is received by the other, downstream sensor #2 (left side). However, propagation between the air and metal pipe wall is very difficult due to mismatching of acoustic impedance. Thus, focusing method of ultrasound is developed. Rounded surface of ultrasonic sensor makes focused ultrasound and increases the signal-to-noise ratio (S/N ratio).



Fig. 2. Ultrasound path with focused sensor.

In the focusing sensors, ultrasonic beam spread is prevented and high amplitude and SNR signal are able to be detected. To verify the effect of focusing sensors, the preliminary experiment is perform, and Fig. 7 shows the experimental apparatus that consists of the ultrasonic sensors, three axis stage, acrylic pipe fulfilled with water, ex-amplifier, pulser/receiver(Japan Probe, JP-10CN), and personal computer. The pulser emits the burst signal to one of the sensor, and the electric signals are converted to ultrasound. The propagated ultrasound is detected on the other sensor, amplified 60 dB on ex-Amplifier, and transfer to the receiver. In the receiver, the signal is converted to digital data and recorded in the PC. The distance between sensors and the pipe wall are 10 mm, and tested materials of pipes are acryl, aluminum alloy, and stainless steel. The tested sensors are focusing sensors (Japan Probe Focusing $0.4K20 \times 20N40R$) and planar sensors (Japan Probe 0.4K20 \times 14). The center frequencies of the sensors are 360 kHz. Transmitted signal is 200 V, two waves burst signal.

The focus ultrasonic sensors (Fig. 3) were validated with horizontally flowing water in a pipe, and showed its applicability for it. Further studies are proceeded for measurement of steam flowrate.



Fig. 3. The developed focusing sensor (right) and a conventional planar sensor (left).

Reference

1. K. Tsukada, et al.; 12th International Conference on Fluid Control, Measurement and Visualization (FLUCOME 2013), 18-23, November, Nara, OS14-4-1 (2013).

C.5

Study on Flow Behavior with Joule Heating

Hiroshige Kikura and Nobuyoshi Tsuzuki

1. Introduction

High-Level radioactive Waste (HLW) has been produced with the operation of nuclear power plant or related facilities, and Liquid-Fed Ceramic Melter (LFCM) has been applied in Japan to make vitrified glass for geological disposal. The LMCF melts glass beads and High-Level radioactive Liquid Waste (HLLW), which is solution of HLW by acid, by electric power as Joule-heating. A test-type melter and a mockup model of LFCM were already fabricated, however, the operation of the melter was not so easy. There are many factors in the melter which affect flow behavior of molten glass. These factors are: convection generated by Joule-heating, effect of electro-magnetic field, shape of the melter, existence of platinum group metal, cooling for electrodes, heat radiation, 'cold cap layer' in which glass beads and other ingredients are melting and gas is generated by chemical reaction, and physical properties of glass itself also affect the glass flow behavior. Therefore, understanding the flow behavior in the melter is very difficult. When sudden change occurred in the glass melter, the operation of the melter must be shut down because estimation of the reason and result from the sudden change is very difficult. Consequently, study on flow behavior in the glass melter has been required and several projects are progressing.



Simplification is necessary to clarify this very complicated flow behavior, thus, a cubic cavity is applied as a cavity shape, and glycerol-water solution is applied as working fluid on behalf of molten glass due to its applicability. Thus, flow behavior is studied using a cubic cavity and glycerol as working fluid.

2. Experimental model

Flow behavior in the cubic cavity (Fig. 2) has been studied changing boundary conditions: electrode cooling, top surface cooling and cooling area of top surface, modeling heat radiation and area change of cold cap. Flow behavior profiles in time were obtained by Ultrasonic Velocity Profiler (UVP) method.

UVP method can obtain time-dependent velocity profiles in function of distance from the sensor, however, UVP method is affected by temperature in the media because it uses sound speed to analyze and the sound speed depends on the temperature. Thus, the effect of temperature distribution, which is largest in the vicinity of the heat sink, was revealed using numerical results and physical properties of the working fluid. The maximum temperature difference in the measurement line is $<20^{\circ}$ C and from the temperature dependency of the sound speed, error from the temperature difference is found $\sim3\%$. It is enough small to obtain the velocity profiles, thus, it is found that UVP method is applicable for velocity profile observation for this Joule-heating apparatus.



Fig. 2. Test apparatus.

3. Boundary conditions

Observations of the flow behavior under this Joule-heating condition were executed changing boundary conditions: electrode cooling condition, top surface cooling temperature and area of the top surface cooling. Normal condition of this apparatus makes very chaotic flow behavior as shown in Fig. 3 due to downflow from top surface to the bottom. However, boundary condition change which makes strong downflow sometimes obstructs the downflow and it restricts the area of chaotic flow only in the upper part of the cavity. Other flow characteristics in this Joule-heating flow and boundary conditions were revealed in this study, and it will contribute to the stable operation of the glass melter.



Reference

1. T. T. Duong, et al.; Nuclear Science and Technology, Vol. 3, No. 3, pp. 31-40 (2013).

C.6

Total View for the Treatment of Fukushima Accident

Hiroshige Kikura, Nobuyoshi Tsuzuki and Kazumi Kitayama

1. Introduction

A huge earthquake occurred on 11th, Mar., 2011, and many nuclear plants were damaged by the earthquake, that is so-called, higashi-nihon daishinsai. Fukushima Dai-ichi Nuclear power plant (1F) got especially large damage, and still decontamination or decommissioning in Fukushima is proceeding. To decommission of the reactor and their debris, many idea of plans how to decontaminate or decommission are proposed from many companies or institutes. Most of the plans or schedules are from the aspects of their subject or interest, however, total management and consideration for unexpected things or accidents are important and necessary for decommissioning without delay.

Tokyo TECH built a project for the decommissioning of Fukushima in 2014, and it includes various aspect including treatment and deposition of the Fukushima debris. However, considering total view for the treatment and deposition from the damaged 1F plants are very important. Consequently, total view of the important subjects for the decommissioning is summarized and consideration for the smooth progression will be discussed.

2. Main processes for decommissioning

2.1. Grasping current situation of debris

Before the treatment for the debris or the storage, we must cut the debris into several parts and insert them using some containers to bring. However, we must avoid the criticality of the debris parts, thus, we must investigate the current status of the debris. Information we can obtain from the debris is considered as following now: ingredients, shape, amount, radiation level and distribution (location) of the debris (Fig. 1). To know these information, we must pay as much effort as possible. Ingredient can be estimated from nuclear reaction and used time of core, and amount can also be estimated from the shape and total amount equipped before the accident. Besides, Ultrasonic technique can be used to investigate the shape of debris. Measurement of shape by ultrasound was already examined during the treatment of TMI-2 accident. However, progressed ultrasonic method can be applied for Fukushima's treatment. Shape of debris and flow behavior around the debris can be obtained by Ultrasonic Velocity Profiler method (UVP method), and the flowing information can be used to estimate the leakage point from the vessel or others. The information about the leakage point is also important to apply submerge method during the treatment for less radiation exposure.

2.2 Storage

When we store the parts of debris, we must care about: criticality of the debris, release of heat or radioactive materials from the debris, nuclear safety, and total amount is also important to store.

Criticality must be avoided absolutely, and heat release should be reduced for the prevention from burning injure or fire protection. Nuclear safety and release of radioactive materials should be cared for less radioactive exposure and terrorism. All for these information should be cared and then we can treat all the debris of Fukushima's correctly.

2.3 Treatment and Disposal

To treat or dispose the debris, we must bring them from the plant site to the treatment and/or disposal sites. However, the condition, 'Bring polluted and mixed radioactive material which ingredient is not accurately known' has not been considered, and no law can be applied for like such transportation. We must decide to make a new law for this special condition, or to modify presenting law into applicable one to this condition.

3. Summary

A total view for Fukushima decommissioning is shown and important things for each stage – investigation, storage, treatment & disposal – are listed. For the smooth progression, we must consider the things without exception, and care about other unexpected things which will occur by accident.



Fig. 1. Main processes for the decommissioning.

1. K. Kitayama et al.; Denki-Jouhou, April, pp. 32-46 (2013).

Reference

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C.7

Passive Gamma Spectrometry of Low-Volatile FPs for Accountancy of Special Nuclear Material in Molten Core Material of Fukushima Daiichi Nuclear Power Station

Hiroshi Sagara

INTRODUCTION

For the nuclear material accountancy of molten core material in Fukushima Daiichi unit 1, 2 and 3(1F1, 1F2 and 1F3), feasibility studies of variety of technologies are being performed. As one of the technologies, feasibility study of passive gamma spectroscopy of low-volatile fission products (FPs) for nuclear material accountancy in molten core material has been performed with reviewing TMI-2 experience, and the correlation of actinides and FPs inventory in core of 1F1, 1F2 and 1F3, considering the sensitivity of axial neutron spectrum, void, burnup, enrichment distribution unique to BWR fuel, and also leakage gamma-ray from molten core materials sphere model. The present paper evaluates the applicability of passive gamma spectrometry to the molten core material of 1F, focusing on the low-volatile FPs and SNM inventory analysis and the fundamental characteristics of gamma-rays from fuel debris. And also the numerical simulation of leakage gamma-ray from molten core materials in hypothetical canister is dealt for determination of radioactivity of low-volatile high-energy emission FPs, could be utilized for sensitive which nuclear material(SNM) quantity estimation coupling with SNM/FPs ratio derived from core inventory calculation.

METHODOLOGY

Figure 1 shows the assumed data flow of passive gamma scanning for SNM quantification inside fuel debris. In SNM estimation in fuel debris by passive gamma spectrometry, four data processing steps are needed: (i) measurement of the intensity and energy spectrum of the gamma-rays from the surface of the fuel debris by a gamma-ray detector, (ii) estimation of gamma-emitting radioactivity from high-energy low-volatile FPs with self-attenuation correction, (iii) burnup estimation using the results of gamma and source term FP/SNM spectrometry ratio determination by inventory calculation, and (iv) quantification of the SNMs in the fuel debris using the data obtained in (ii) and (iii).

Figure 2 shows the geometry of "Fuel canister" utilized in molten core material recovery process of Three Mile Island unit 2(TMI-2). In the present paper, the model of TMI-2 canister is used for gamma-ray leakage calculation, with 3 main geometrical regions; canister, surrounding water/air and shielding/collimator for detectors. For the gamma-ray leakage/shielding calculation, MCNP5 code with photon library of

MCPLIB04 was utilized. Homogeneous loading model of molten core material and water/air is taken as a reference model as same as TMI-2 core bore cases, and patterns of heterogeneous loading model are also evaluated, and the effectiveness of several attenuation correction techniques is compared.



Fig. 1 Data flow of passive gamma scanning for SNM quantification.



Fig. 2 TMI-2 fuel canister model: (left) horizontal cross-section, (right) vertical cross-section

RESULTS

Figure 3 shows the horizontal geometry of canister with homogeneous and heterogeneous loading pattern of molten core material; black square suggests molten fuel material block and ivory-white square suggests water block.

Composition of average molten core material is of 1F2 with fuel and zircalloy(HM+FP oxide:Zr=1:1 in weight, 6g/cc, packing fraction: 60vol.%), fulfilled with water inside the canister. Table 1 shows the results of leakage photons emitted from molten core material. Since the low energy gamma-ray of 59.5keV from Am-241 is easier to be shielded, its leakage ratio is very sensitive to the surface material. On the other hand, high energy gamma-rays of 1.28 and 2.71 MeV from Eu-154 and Rh-106 are more transparent and the difference of the material layout with 2-3cm interval has no big sensitivity, and self-attenuation correction by average density homogeneous material model can be applied to this kind of circular symmetric case. More parameters of packing fraction, porosity and composition of molten core material, geometrical heterogeneity are studied and results are summarized as sensitivity to the homogeneous model. Figure 4 shows an example of the calculation result of photon detection spectrum from the fuel canister, comparing the different detectors: a high purity germanium (HPGe) detector and a LaBr₃ scintillation detector with assumed energy resolution of 0.5% and 2.5% respectively. Gamma-ray source in the calculation is a fuel debris from 1F2 core with structure material loaded into the fuel type canister with packing fraction of 60 vol.%, and detection spectrum is calculated by photon transport calculation with detection efficiency data and with Gaussian spectrum adjustment. The result shows that primary and other intensive photo-peaks of Ce/Pr-144 and Eu-154 can be distinguished by both detector types, these could be utilized for the gamma spectrometry of fuel debris in the aspect of energy resolution. The present study also compared several attenuation correction techniques and evaluated the effectiveness.



Fig. 3 A example of horizontal cross-section of fuel canister; (left)homogeneous model, (right)heterogeneous model.

Table	1 I	eakage	ratio	of s	necific	energy	gamma-ray	
Iunic		Jounugo	iuuo	OI D	peenie	Unor S y	Samma ray	

-	Commo oporqu(Nuolido)	Leak Photon/Source Photon			
	Gamma energy(Nuclide)	Homo	Hetero		
	59.5keV(Am-241)	6.64E-07	3.44E-07		
	1.28MeV(Eu-154)	8.66E-02	8.37E-02		
-	2.71MeV(Rh-106)	1.51E-01	1.48E-01		



Fig. 4 An example of predicted photon detection spectrum by different detectors. (Assumed fuel debris: 1F2 heavy metal with 10-year cooling loaded in fuel type canister)

Figure 5 shows images of the passive gamma scanning system. The module package system is assumed to be installed in a storage pool in the reactor building or in the common storage facility at the 1F site, with adequate shielding, a collimator, and equipment such as a crane. To correct for self-shielding and the heterogeneity of the photon source inside the fuel debris and canister, the applicability of several techniques would be considered, such as vertical rotation/face to- face and axial probing scans; attenuation coefficient estimation using the correlation with the leakage photopeak ratio, average density from the weight, and external transmission source; tomography technology with passive gamma or active photons; and/or the sample characteristics in each stratum assayed by DAs and NDAs in laboratories, as applied to TMI-2. The type of detector and collimator design must also be selected according to the requirements for resolution, operation time, and efficiency. A high purity germanium detector is a practical candidate, and portable scintillation detectors such as NaI or LaBr3 might also be worth surveying for the canister or for areas where the accessibility to residual fuel debris would be difficult, such as in cooling pipes, as applied in TMI-2. At Tokyo Institute of Technology and the Japan Atomic Energy Agency, passive gamma measurement of irradiated fuel or radioisotopes is being examined experimentally to obtain a correlation table for self-shielding and heterogeneity correction. The FP release fractions are also being surveyed the historical experimental data collection and hv measurement in mockup debris of spent nuclear fuel before and after melting, and the results will be used to determine the range of low-volatile FP release fraction and to ensure the coexistence of FPs with SNMs.



Fig. 5 Image of the passive gamma measurement module (in air or under water). Notes: RB: reactor building; CSFP: common spent fuel pool.

REFERENCES

- 1. H.Sagara et al., J. Nucl. Sci. Technol., vol.51, no.1, pp. 1-23 (2014).
- 2. H.Sagara et al., Trans. Am. Nucl. Soc., Vol. 108, pp. 2316-2317 (2013).

II. Co-operative Researches

II. Co-operative Researches

II.1 Co-operative Researches within Tokyo Institute of Technology

Under the generic proposition: "The Release and Utilization of Energies existing in Atoms, Molecules and Nuclei" which has been hoisted since the reorganization of RLNR, we advertise for collaborations with any department and laboratory within Tokyo Institute of Technology in order to develop researches for the improvement of energy utilization with safety, mass transmutation by nuclear reactions with high efficiency, and construction of energy system concept with high societal acceptability. The RLNR will promote these collaborations by offering the organizations, facilities, equipments and space as the research base for research project teams and research groups crossing over within Tokyo Tech. We are going to adopt unique and innovative themes which make maximum use of the activities of this research laboratory.

• Development of the Clamp-on Ultrasound Flow Meter for Steam in Pipe Department of Mechanical Engineering, Tokyo Institute of Technology

II.2 Co-operative Researches with Outside of Tokyo Institute of Technology

- Emission Spectroscopic • Research on Optical Measurement of High-Density Helicon Plasma for Application to Electric Propulsion Rocket Tokyo University of Agriculture and Technology
- Observation of Optical Emission Spectroscopy in Atmospheric-pressure Plasma Jet University of Miyazaki
- Innovative characterization of materials under severe condition

Centre National de la Recherche Scientifique

- Structure and physico-chemical properties of actinides fluorides Tohoku University
- XAFS analysis and UV-vis spectroscopy of halides containing actinide and fission products Kyoto University
- Treatment of fuel debris using selective fluorination and molten salt electrolysis, Grant-in-Aid for Challenging Exploratory Research Japan Society for the Promotion of Science

• Experimental investigation of positional stabilization of

torus plasma with helical coils National Institute for Fusion Science

- Development of Fiber-Optic Diagnotic on Vacuum Vessel Current of OUEST Kyushu University
- Study on Diagnostics Applicable to Basic Control of **DEMO** Fusion Reactors Japan Atomic Energy Agency
- n TOF Collaboration CERN
- A Study on Separation of Uranyl Species Using Alginic Acid

Assiut University, Egypt

• Studies on Separation of Metal Species Using Ionic Liquids Laboratoire d'Electrochimie et de Physicochimie des

Matériaux et des Interfaces, France

- Ion Beam Analysis of Fuel-Cell Materials, Department of Chemistry and Energy Engineering, Faculty of Engineering Tokyo City University
- Astrobiology Experiments Based on MeV Ion Beams, Division of Materials Science and Chemical Engineering, Faculty of Engineering Yokohama National University
- Development of Negative Ion Sources for Tandem Accelerators, Atomic Energy Research Laboratory Tokyo City University
- Selective Irradiation of Cancer Cells by Electrons Emitted from Cancer-Localizing Drugs, Grant-in-Aid for Scientific Research (B) Japan Society for the Promotion of Science
- Needle-Shaped Monochromatic Brachytherapy X-Ray Source for Selective Irradiation of Tumors Using X-Ray Absorption Characteristics of Drugs, Grant-in-Aid for Challenging Exploratory Research Japan Society for the Promotion of Science
- High-Brightness Monochromatic Micro X-Ray Source Based on Capillary X-Ray Optics, Grant-in-Aid for Scientific Research (B) Japan Society for the Promotion of Science
- Study on Double Decay of ⁴⁸Ca Osaka University

• Study on the possibility of ultra-high burnup core and fuel

Japan Atomic Energy Agency

- Study on the possibility of CANDLE reactor Central Research Institute of Electric Power Industry
- High-Efficient Utilization of Medium-Temperature Heat by Using High-Thermal Conductivity Materials, Grant-in-Aid for Scientific Research (B) Japan Society for the Promotion of Science
- Load-Leveling of Renewable Energy System by Using Direct Carbon Dioxide Electrolysis, Grant-in-Aid for Exploratory Research Japan Society for the Promotion of Science
- Development of materials for chemical heat storage with metal organic framework (MOF) as host matrix, Grant-in-Aid for Exploratory Research Japan Society for the Promotion of Science
- Thermochemical energy storage material and system for high-temperature heat storage Japan Science and Technology Agency
- Development of materials for ammonia separation and storage
 - Japan Science and Technology Agency
- Collaborative research on thermal-hydrodynamics for future light water reactor, energy system and chemical technology development Vietnam Academy of Science and Technology
- Collaborative research on two-phase flow dynamics for future light water reactor development Korea Atomic Energy Research Institute
- Collaborative research on fluid engineering Muroran Institute of Technology
- Collaborative research on multiphase flow engineering Nihon University
- Collaborative research on treatment, disposal of radioactive waste and nuclear safety Okayama University
- Collaborative research on thermal-hydrodynamics for future light water reactor, energy system and chemical technology development Swiss Federal Institute of Technology
- Development of the Removal Process of Platinum Group Metals from High-level Radioactive Waste for attaining high quality and volume reduction for the

vitrified object Japan Science and Technology Agency

- Fundamental Test Related with Physical Property of Glass -Advancement of Vitrification process-Japan Nuclear Fuel Limited
- Research on Thermo-decomposition / Volume Reduction of Ferric Ferrocyanide Absorbed Cesium (2) Maeda Corporation
- Development of Functional Sheet for the Removal of Platinum Group Metals and Heat-generating Nuclides from High-level Radioactive Waste Nitto Denko Corporation.
- Development of Absorbents for Decontamination from radioactive contamination Nitto Denko Corporation.
- Basic Studies for Developing Rational Treatment and Disposal System of Radioactive Wastes Generated by Fukushima Nuclear Reactor Accident, Grant-in-Aid for Scientific Research (S) Japan Society for the Promotion of Science
- Development of Liquid-liquid Counter-current Centrifugal Extractor for Nuclide Separation, Grant-in-Aid for Scientific Research (B), Japan Society for the Promotion of Science
- Development of the Gel-Liquid Extraction Separation Process for Elimination of Radioactive Cesium and Strontium, Grant-in-Aid for Young Scientists (B) Japan Society for the Promotion of Science
- Development of A Novel Nanofluidic-based Separation System for Rare-Earth Elements e, Funding Program for Next Generation World-Leading Researchers (NEXT Program)

Japan Society for the Promotion of Science

- Development of an atmospheric scanning positron microscope National Institute of Advanced Industrial Science and Technology
- Advanced study on heavy ion linac National Institute of Radiological Sciences
- Regulation of DNA repair and cell death through phosphorylation of DNA repair proteins Kanazawa Medical University
- Recovery of Neutron-Induced Defects and Release of Helium Gas using Nano-Dilatometry Equipped with Mass Spectroscopy, Grant-in-Aid for Scientific

Research

Japan Society for the Promotion of Science

• Recovery of Neutron-Induced Defects and Release of Helium Gas using Nano-Dilatometry Equipped with Mass Spectroscopy, Grant-in-Aid for Scientific Research

Japan Society for the Promotion of Science

- Study on Low-Cost Process of SiC/SiC Composites Japan Aerospace Exploration Agency
- Study on Properties of B₄C Neutron-Absorbing Materials for Control Rods Japan Atomic Energy Agency
- Study on Neutron-Irradiation Resistance of Orientation-Controlled Ceramics National Institute for Materials Science
- Sinterability of SiC Ceramics with Al₄SiC₄ Addition and Their Properties National Institute for Materials Science
- Study on the Evaluation of Thermal Shock Fracture Behavior of Ceramics Tokyo Metropolitan University
- Research on Formation and Characterization of Oxide Nanopowder University of Belgrade, Serbia
- Passive Gamma Spectrometry of Low-Volatile FPs for Accountancy of Special Nuclear Material in Molten Core Material of Fukushima Daiichi Nuclear Power Plant

Japan Atomic Energy Agency

• Strong magnetic field generation using high intensity laser

Institute of Laser Engineering, Osaka University

• Ion charge state analysis in tantalum plasmas using a pico-second laser, FY2013 Research Abroad and Invitational Program for the Promotion of International Joint Research

Collider Accelerator Department, Brookhaven National Laboratory

III. List of Publications

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Yoh Nagahara, Hiroki Ichii, Kazuyuki Yoshida, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akatsuka: Characteristics of Cold Argon Arc-Jet Plasma Flowing Along Open-Field-Line and the Effects of Collisions on Deceleration; *IEEE Transactions on Plasma Science*, Vol.**41**, No.8, pp. 1869-1877 (2013).

H. Akatsuka, M. Hatcho, H. Matsuura: Nitrogen Arc-Jet as Atomic Radical Source - Dominance of Atomic N Lines in Comparison with Molecular N₂ Band Spectra; *The 12th Asia Pacific Physics Conference, APPC12 Abstracts, (APPC12)* July 15-19, 2013, Chiba, Japan, p. 989, JPS Conf. Proc. Vol.1, 015079 (2014) [4 pages].

H. Akatsuka: Discussion on Collisional Radiative Model from the Viewpoint of Linear Ordinary Differential Equations; *The 12th Asia Pacific Physics Conference, APPC12 Abstracts, (APPC12)* July 15-19, 2013, Chiba, Japan, p. 579, *JPS Conf. Proc.* Vol.1, 015016 (2014) [4 pages].

Yuki Ohno, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akasuka: Spectroscopic Study of Recombination Continuum in Arc-Heated Cold Expanding Plasma Jet; *Program and Abstracts of The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Nov. 6-8, 2013, Tokyo, Japan, P-09.

Yoshihiro Shimizu, Yuusuke Kittaka, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akatsuka: Excited State Distributions of Hydrogen Atoms in Microwave Discharge Hydrogen Plasma and Effect of Electron Energy Distribution Function; *Program and Abstracts of The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Nov. 6-8, 2013, Tokyo, Japan, P-10.

Satoshi Tsuno, Ryutaro Yamashiro, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akatsuka: Numerical Study on Acceleration and Deceleration Mechanisms of Weakly Ionized Plasma Flowing Supersonically through Open Field Lines; *Prog. 8th Asia-Pacific International Symposium on the Basics and Application of Plasma Technology (APSPT-8)*, Dec. 20-22, 2013, Hsinchiu, Taiwan, p. 196.

Hiroshi Akatsuka, Masahiko Hatcho, Haruaki Matsuura: Spectroscopic Characteristics of Supersonically Expanding Nitrogen Arc-Jet Plume — Strong Atomic Lines and Weak Molecular Band Spectra; 8th Asia-Pacific International Symposium on the Basics and Applications of Plasma Technology, *Prog. 8th Asia-Pacific International Symposium on the Basics and Application of Plasma Technology (APSPT-8)*, Dec. 20-22, 2013, Hsinchiu, Taiwan, p. 121.

Hao Tan, Atsushi Nezu, Haruaki Matsuura, Hiroshi

Akatsuka: Calculation and Spectroscopic Examination of γ -band Spectra of NO Molecule by Using Microwave Discharge N₂-O₂ Mixture Plasma; *Proc. 8th International Conference on Reactive Plasmas/31st Symposium on Plasma Processing (ICRP8/SPP31)*, Feb. 3-7, 2014, Fukuoka, Japan, 5B-PM-O5.

Hiroshi Akatsuka, Yoshihiro Shimizu, Atsushi Nezu, Haruaki Matsuura: Dependence on Vibrational Quantum Number of H₂ Rotational Temperature of a Low-Pressure Microwave Discharge Plasma; *Proc. 8th International Conference on Reactive Plasmas/31st Symposium on Plasma Processing (ICRP8/SPP31)*, Feb. 3-7, 2014, Fukuoka, Japan, 5P-AM-S02-P01.

Junya Konno, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akatsuka: Excitation Kinetics of Discharge Oxygen Plasma with Low Pressure and Effect of EEDF; *The Papers of Joint Technical Meeting on "Plasma Science and Technology", "Pulsed Power Technology" and "Electrical Discharges", IEE Japan, May 16-17, 2013, Nagaoka, Japan, PST-13-063, PPT-13-048, ED-13-053, pp. 23-28.*

Hiroshi Akatsuka, Toshifumi Yuji, Narong Mungkung, B. W. Park, Y. M. Sung: Plasma Treatment of TiO₂ Films for Dye-Sensitized Solar Cells by Non-Equilibrium Plasma Jet with Atmospheric Pressure; *Abstracts of 11th Annual Meeting of Society of Nano Science and Technology*, Jun. 6-8, 2013, Tokyo, Japan, P1-48, p. 138.

Masahiko Hatcho, Sou Watanabe, Haruaki Matsuura, Hiroshi Akatsuka: Peculiarity of Optical Emission Spectroscopic Characteristics of Expanding Nitrogen Arc-Jet Plasma; *Proc. Space Transportation Symposium: FY2012*, Jul., 2013, STEP-2012-029.

Hiroshi Akatsuka, Yoshihiro Shimizu, Atsuhi Nezu, Haruaki Matsuura: Dependence on Vibrational Quantum Number of H₂ Rotational Temperature of a Low-Pressure Microwave Discharge Plasma; *Extended Abstracts of the 74th Autumn Meeting, 2013*, Sept. 16-20, 2013, Kyotanabe, Japan, 17p-P2-8.

Hao Tan, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akatsuka; Calculation of γ -band Spectra of NO Molecule in Microwave Discharge N₂-O₂ Mixture Plasma and Determination of Vibrational and Rotational Temperature; *The Papers of Technical Meeting on "Plasma Science and Technology", IEE Japan*, Sept. 5-7, 2013, Nagasaki, Japan, PST-13-104-113, pp. 29-32.

Satoshi Tsuno, Hiroshi Akatsuka: Numerical Study on Movement of Ion and Neutral Atoms of Weakly Ionized Plasma Flowing Supersonically through Open Field Lines; *The Papers on Technical Meeting on "Plasma Science and Technology", IEE Japan*, Nov. 21-23, 2013, Nagoya, Japan, PST-13-138, pp. 39 - 42. Hiroaki Fujitsuka, Simpei Waseda, Marie Sakata, Daisuke Kuwahara, Hiroshi Akatsuka, Shunjiro Shinohara: Study on Completely Electrodeless Electric Propulsion System Using High-Density Helicon Plasma (IV): 2D Imaging by Spectroscopic Method; *On-Line Abstracts of the 30th Annual Meeting of Japan Society of Plasma Science and Nuclear Fusion Research*, Dec. 3-6, 2013, Tokyo, Japan, 03aE20P.

Junya Konno, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akatsuka: Effect of EEDF on the Excitation Kinetics of Low-Pressure Discharge Oxygen Plasma; *On-Line Abstracts of The 30th Annual Meeting of Japan Society of Plasma Science and Nuclear Fusion Research, Japan Society of Plasma Science and Nuclear Fusion Research, Dec.* 3-6, 2013, Tokyo, Japan, 03aE12P.

Satoshi Tsuno, Takeshi Nakahagi, Atsushi Nezu, Haruaki Matsuura, Hiroshi Akatsuka: Hybrid Simulation of Supersonic Rarefied Flow of Weakly Ionized Plasma along Open Field Magnetic Line; 2014 Annual Meeting Record of IEE Japan, Mar. 18-20, Matsuyama, Japan, p. 147, 1-128.

J. Konno, A. Nezu, H. Matsuura, H. Akatsuka: Essentiality of EEDF on the Excitation Kinetics of Low-Pressure Discharge Oxygen Plasma; *Meeting Abstracts of the Physical Society of Japan*, Mar. 27-30, Hiratsuka, Japan, Vol.**69**, No.1, p. 282, 30aAX-13.

Hiroo Numata, Hiroshi Akatsuka and Haruaki Matsuura: Optimization of a Molten Salt Electrolyte Bath Geometry for Rare Earth Metal Recovery using a Finite Element Method; *Z. Naturforsch.*, **68**a, 48-58 (2013).

Haruaki Matsuura, Atsushi Nezu, Kazuyuki Yoshida, Hiroshi Akatsuka, Akihiko Kajinami, Jun-ichi Nishino, Kentaro Kajiwara, Norimasa Umesaki, Akihiro Kawabe: In-situ observation of electrochemical deposition of rare earth in molten chlorides by using X-ray from synchrotron source; *Abstract of The 4th International symposium on Innovative Nuclear Energy Systems*, November, Tokyo, Japan, B13-4, 2013.

Kazuhiro Fujita , Satoshi Tsuno, Ryutaro Yamashiro, Toshiyuki Fujii, Hajimu Yamana, Atsushi Nezu, Hiroshi Akatsuka and Haruaki Matsuura, : Fluoride addition effect on electrochemical behavior and local structure around neodymium cation in molten LiCl-KCl eutectic; *Proceedings of the 45th Symposium on Molten Salt Chemistry*, November, Yokohama, Japan, 9-10, 2013.

Haruaki Matsuura, Atsushi Nezu, Hiroshi Akatsuka, Hideaki Shiwaku, Tsuyoshi Yaita, Hajimu Yamana, Toshiyuki Fujii, Akihiro Uehara: Effect of alkali or alkaline earth cation on local structure around neodymium cation in molten chlorides; *Proceedings of the autumn conference of the* *electrochemistry society of Japan*, September, Tokyo, Japan, 283, 2013.

Kazuhiro Fujita, Atsushi Nezu, Satoshi Tsuno, Ryutaro Yamashiro, Hiroshi Akatsuka, Hajimu Yamana, Toshiyuki Fujii, and Haruaki Matsuura : Fluoride addition effect on local structure of neodymium cation in molten LiCl - KCl; *Proceedings of the 80th memorial conference of the electrochemistry society of Japan*, March, Sendai, Japan, 340, 2013.

S. Maeyama, A. Ishizawa, T.-H. Watanabe, N. Nakajima, S. Tsuji-Iio, H. Tsutsui: Numerical techniques for parallel dynamics in electromagnetic gyrokinetic Vlasov simulations; *Comput. Phys. Commun.* Vol. **184**, pp. 2462-2473 (2013).

S. Hatakeyama, M. Miura, Z. Yao, H. Tsutsui, S. Tsuji-Iio, Y. Shibata, N. Ohno, K.Y. Watanabe, T. Akiyama, Y. Suzuki, K.Y. Watanabe: Construction of a Small Tokamak Device with Vertically Positional Stability and with an Elongated Cross Section; *2013 Autumn Meeting of Physical Society of Japan*, Tokushima, Sep. 25-28, 2013, 28pKB-1.

Tatsuya Katabuchi, Brian Hales, Noriyosu Hayashizaki, Masayuki Igashira, Zareen Khan, Tooru Kobayashi, Taihei Matsuhashi, Koichi Miyazaki, Koichi Ogawa, Kazushi Terada: Feasibility study on pinhole camera system for online dosimetry in boron neutron capture therapy; *Applied Radiation and Isotopes*, Vol. **88**, pp. 139-142 (2013).

B. Hales, T. Katabuchi, N. Hayashizaki, K. Terada, M. Igashira, T. Kobayashi: Feasibility study of SPECT system for online dosimetry imaging in boron neutron capture therapy; *Applied Radiation and Isotopes*, Vol. **88**, pp. 167-170 (2013).

K. Kino, M. Furusaka, F. Hiraga, T. Kamiyama, Y. Kiyanagi, K. Furutaka, S. Goko, K.Y. Hara, H. Harada, M. Harada, K. Hirose, T. Kai, A. Kimura, T. Kin, F. Kitatani, M. Koizumi, F. Maekawa, S. Meigo, S. Nakamura, M. Ooi, M. Ohta, M. Oshima, Y. Toh, M. Igashira, T. Katabuchi, M. Mizumoto, J. Hori: Energy resolution of pulsed neutron beam provided by the ANNRI beamline at the J-PARC/MLF; *Nuclear Instruments and Methods in Physics Research Section A*; Vol. **736**, pp. 66-74, (2013).

Tooru Kobayashi, Noriyosu Hayashizaki, Tatsuya Katabuchi, Kenichi Tanaka, Gerard Bengua, Noriaki Nakao, Kazuaki Kosako: Near-threshold ⁷Li(p,n)⁷Be neutrons on the practical conditions using thick Li-target and Gaussian proton energies for BNCT; *Applied Radiation and Isotopes*, Vol. **88**, pp. 221-224 (2014).

Kazushi Terada, Taihei Matsuhashi, Masayuki Igashira, Tatsuya Katabuchi: Measurements of keV-neutron capture

cross sections and capture gamma-ray spectra of ¹⁰⁵Pd, *Journal of Nuclear Science and Technology*; Vol. **51**, pp. 671-680 (2014).

Kazushi Terada, Tatsuya Katabuchi, Masayuki Igashira, Taihei Matsuhashi, Tuan Anh TRAN: Systematic Measurements of keV-Neutron Capture Cross Sections and Capture Gamma-Ray Spectra of Pd Isotopes, *14th International Symposium on Capture Gamma-Ray Spectroscopy and Related Topics (CGS15)*, Aug 28 – Sept 2, 2011, Guelph, Canada, Proceedings of the 14th International Symposium on Capture Gamma-Ray Spectroscopy and Related Topics, World Scientific, pp. 456-461 (2013).

Tatsuya Katabuchi, Masayuki Igashira, Michihide Tajika, Yuichi Nakamura, So Kamata, Kazushi Terada: Measurement of keV-neutron capture cross section and gamma-ray spectra of ¹⁴²Nd, *14th International Symposium on Capture Gamma-Ray Spectroscopy and Related Topics (CGS14)*, Aug 28 – Sept 2, 2011, Guelph, Canada, Proceedings of the 14th International Symposium on Capture Gamma-Ray Spectroscopy and Related Topics, World Scientific, pp. 238-243 (2013).

Brian Hales, Tatsuya Katabuchi, Noriyosu Hayashizaki, Kazushi Terada, Masayuki Igashira, Tooru Kobayashi: Shielding Design of Prompt-Gamma SPECT for Boron Neutron Capture Therapy; 2013 Fall Meeting of the Atomic Energy Society of Japan, September 3-5, 2013, C22.

K. Teraeda, T. Katabuchi, M. Mizumoto, T. Arai, T. Saito, M. Igashira, K. Hirose, S. Nakamura, A. Kimura, H. Harada, J. Hori, K. Kino, Y. Kiyanagi: Measurements of neutron capture cross section of Pd-107 with an NaI(Tl) spectrometer at J-PARC/MLF/ANNRI, 2013 Fall Meeting of the Atomic Energy Society of Japan, September 3-5, 2013, C01.

A. Kimura, K. Hirose, S. Nakamura, K. Hara, H. Harada, G. Chiba T. Katabuchi, K. Terada, M. Mizumoto, M. Igashira: Measurement of neutron capture cross section of Gd-157 with the NaI(Tl) spectrometer and the Ge spectrometer in J-PARC/MLF/ANNRI, *2013 Fall Meeting of the Atomic Energy Society of Japan*, September 3-5, 2013, C02.

T. Arai, T. Katabuchi, K. Terada, M. Igashira, A. Kimura, H. Harada, S. Nakamura, J. Hori, K. Kino, Y. Kiyanagi: Measurement of Neutron Capture Cross Section of Am-241 using ANNRI-NaI(TI) Spectrometer at J-PARC, 2014 Annual Meeting of the Atomic Energy Society of Japan, March 3-5, 2014, J08.

B. Machura, M. Wolff, E. Benoist, J.A. Schachner, N.C. Mösch-Zanetti, K. Takao, and Y. Ikeda: Oxorhenium(V) Complexes with 1*H*-Benzimidazole-2-carboxylic acid –

Synthesis, Structural Characterization and Catalytic Application in Epoxidation Reactions; *Polyhedron*, **69**, 205-218 (2014).

T. Suzuki, K. Takao, T. Kawasaki, M. Harada, M.Nogami, and Y. Ikeda: A Sudy on Selective Precipitation of U(VI) by Hydrophilic Cyclic Urea Derivatives for Development of a Reprocessing System Based on Precipitation Method; *J. Nucl. Sci. Technol.* **51**, 514-520 (2014).

T. Kawasaki, S. Okumura, Y. Sasaki, and Y. Ikeda: Crystal Structures of Ln(III) (Ln = La, Pr, Nd, Sm, Eu, Gd) Complexes with N, N, N', N'-Tetraethyldiglycolamide associated with homoleptic $[Ln(NO_3)_6)]^3$; *Bull. Chem. Soc. Jpn.*,**87**, 294-300 (2014).

K. Sasaki, K. Takao, T. Suzuki, T. Mori, T. Arai, and Y. Ikeda: Extraction of Pd(II), Rh(III) and Ru(III) from HNO₃ Aqueous Solution to Betainium Bis(trifluoromethanesulfonyl)imide Ionic Liquid; *Dalton Trans.*, **43**, 5648-5651 (2014).

K. Sasaki, T. Suzuki, T. Arai, K. Takao, S. Suzuki, T. Yaita, and Y. Ikeda: Uranyl Species in 1-Ethyl-3-methylimidazolium Nitrate $([EMI][NO_3])$ [EMI]- $[UO_2(NO_3)_4]$ Solution of First Specrophotometric Evidence Existance for of $[UO_2(NO_3)_4]^{2-}$; Chem. Lett., 43, 670-672 (2014).

Y. Ohashi, M. Nomura, Y. Tsunashima, S. Ando, N. Sugitsue, Y. Ikeda, and Y. Tanaka: Technique for Revovering Uranium from Sludge-like Uranium-bearing Wastes Using Hydrochloric Acid; *J. Nucl. Sci. Technol.*, **61**, 251-265 (2014).

K. Sasaki, T. Suzuki, T. Mori, T. Arai, K. Takao, and Y. Ikeda: Selective Liquid-Liquid Extraction of Uranyl Species Using Task-Specific Ionic Liquid, Betainium Bis(trifluoromethylsulfonyl)imide; *Chem. Lett.*, **43**, 775-777 (2014).

K. Teramoto, T. Nishide, S. Okumura, K. Takao, and Y. Ikeda: Studies on Metal Complexes as Active Materials in Redox-flow Battery Using Ionic Liquids as Electrolyte: Electrochemical Properties of $[Fe(L)_x][Tf_2N]_2$ (L: multidentate ligands, x = 2 or 3) in 1-Butyl-3-methylimidazolium Bis (trifluoromethylsulfonyl)imide, $[BMI][Tf_2N]$; *Electochem.*, **82**, 566-572 (2014).

M.C. Ali, T. Kawasaki, T. Suzuki, Y. Sasaki, and Y. Ikeda: Syntheses and Crystal Structures of Eu(III) and Sm(III) Perrhenate Complexes with 2,2'-(Imino)bis (*N*,*N*'-diethylacetamide); *J. Nucl. Sci. Technol.*, **51**, 1133-1140 (2014). K. Takao, S. Tsushima, T. Ogura, T. Tsubomura, and Y. Ikeda: Experimental and Theoretical Approaches to Redox Innocence of Ligands in Uranyl Complexes: What is Formal Oxidation States of Uranium in Reduction of Uranyl(VI)?; *Inorg. Chem.*, **53**, 5772-5780 (2014).

T. Mori, K. Sasaki, T. Suzuki, T. Arai, K. Takao, and Y. Ikeda: Homogeneous Liquid-liquid Extraction of Pd(II), Rh(III), Ru(III), and U(VI) Using a Carboxyl-functionalized Ionic Liquid with Thermomorphic Behavior; 2nd International Conference on Ionic Liquids in Separation and Purification Technology, Toronto, Canada, June 29 – July 2, 2014.

D. Hwang, T. Tsukahara, and Y. Ikeda: Solubility Studies on Lanthanide(III) Fluorinated Complexes in Supercritical Carbon Dioxide; *The 2nd Asian Nuclear Fuel Conference*, Sendai, Miyagi, Japan, Sep. 18-19, 2014.

R. Ito, T. Tsukahara, and Y. Ikeda: Synthesis of Crown Ether Derivative with Fluorinated Arms for Decontamination of Radioactive Wastes Using Supercritical Carbon Dioxide as Medium; *The 2nd Asian Nuclear Fuel Conference*, Sendai, Miyagi, Japan, Sep. 18-19, 2014.

K. Arakawa, K. Takao, and Y. Ikeda: Studies on Extraction and Recovery of U(VI) in HNO_{3aq}-[BMI][NfO] Ionic Liquid Biphasic System for Decontamination of Uranium Wastes; The 2nd Asian Nuclear Fuel Conference, Sendai, Miyagi, Japan, Sep. 18-19, 2014.

Y. Fukatsu, T. Tsukahara, Y. Ikeda: Molecular Structure of Water inside Porous Silica and Its Dynamics by NMR; *94th Spring Meeting of Chemical Society of Japan*, Nogoya, March 27-30, 2014.

T. Watanabe, K. Takaoa, Y. Ikeda: A Study on Adsorption Mechanism of Uranyl Carbonate Complexes onto Tannin Adsorbent; *The Annual Meeting of the Atomic Energy Society of Japan*, Tokyo, March 26-28, 2014.

K. Takao, T. Suzuki, M. Harada, Y. Ikeda: Molecular Structural Studies on Uranyl Nitrate Precipitates Formed by the Reaction with 2-Pyrollidone Derivatives; *The Annual Meeting of the Atomic Energy Society of Japan*, Tokyo, March 26-28, 2014.

D. Hwang, T. Tsukahara, Y. Ikeda: Basic Studies for Developing Rational Treatment and Disposal System of Radioactive Wastes Generated by Fukushima Dai-ichi Nuclear Accident(16) Studies on Solubility of Metal Complexes in Supercritical CO_2 for Developing Decontamination Method by Using Supercritical CO_2 as Media; *The Annual Meeting of the Atomic Energy Society of Japan*, Tokyo, March 26-28, 2014. T. Suzuki, Y. Sasaki, T. Kawasaki, K. Takaoa, Y. Ikeda, M. Harada: Relationship between Crystal Structure of Uranyl Nitrate complexes with Cyclic Urea Derivatives and Their Melting Points; *The Fall Meeting of the Atomic Energy Society of Japan*, Kyoto, September 8-10, 2014.

T. Mori, K. Takao, K. Arakawa, Y. Ikeda, Basic Studies for Developing Rational Treatment and Disposal System of Radioactive Wastes Generated by Fukushima Dai-ichi Nuclear Accident(26) A Study on Decontamination Method Using Ionic Liquids; *The Fall Meeting of the Atomic Energy Society of Japan*, Kyoto, September 8-10, 2014.

Koichiro Takao, Thomas J. Bell, Yasuhisa Ikeda: Actinide Chemistry in Ionic Liquids; *Inorganic Chemistry*, Vol.**52**, pp. 3459-3472 (2013).

Haruka Uesugi, Toshiaki Tsukuda, Koichiro Takao, Taro Tsubomura: Highly Emissive Platinum(II) Complexes Bearing Carbene and Cyclometalated Ligands; *Dalton Transactions*, Vol.42, pp. 7396-7403 (2013).

Christoph Hennig, Shinobu Takao, Koichiro Takao, Stephan Weiss, Werner Kraus, Franziska Emmerling, Michel Meyer, Andreas C. Scheinost: Identification of Hexanuclear Actinide(IV) Carboxylates with Thorium, Uranium and Neptunium by EXAFS Spectroscopy; *Journal of Physics: Conf. Series*, Vol.**430**, p. 012116 (2013).

Taro Tsubomura, Hironori Murota, Koichiro Takao: Luminescent Palladium(0) Complexes Bearing N-Heterocyclic Carbene and Phosphine Ligands; *Inorganic Chemistry Communications*, Vol.**35**, pp. 110-112 (2013).

Koichiro Takao, Shinobu Takao, Yasuhisa Ikeda, Gert Bernhard, Christoph Hennig: Uranyl-Halide Complexation in N,N-Dimethylformamide: Halide Coordination Trend Manifests Hardness of $[UO_2]^{2+}$; *Dalton Transactions*, Vol.**42**, pp. 13101-13111 (2013).

T. Mori, K. Sasaki, T. Suzuki, T. Arai, K. Takao, Y. Ikeda: Separation of Uranyl Species Using Ionic Liquid Having Property of Temperature-dependent Miscibility with Water; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, November 6-8, 2013, Tokyo Japan, B13-2.

K. Sasaki, T. Suzuki, T. Arai, K. Takao, S. Suzuki, T. Yaita, Y. Ikeda: Structural Studies on Uranyl Species in Acetonitrile and 1-Ethyl-3-methylimidazolium Nitrate ([EMI][NO3]) Dissolved [EMI]₂[UO₂(NO₃)₄] –Evidence for the formation of $[UO_2(NO_3)_4]^{2^\circ}$; *The 9th International Conference on the Chemistry and Physics of the Actinide Elements (Actinides 2013)*, July 21-26, 2013, Karlsruhe, Germany, 1-13.
T. Suzuki, T. Kawasaki, K. Takao, M. Harada, M. Nogami, Y. Ikeda: Effect of Hydrogen Bonds on Melting Points and Packing Coefficients of Uranyl Nitrate Complexes with Cyclic Urea Derivatives; *The 9th International Conference on the Chemistry and Physics of the Actinide Elements (Actinides 2013)*, July 21-26, 2013, Karlsruhe, Germany, 3-37.

T. Mori, K. Sasaki, T. Suzuki, T. Arai, K. Takao, Y. Ikeda: Separation of Uranyl Species Using Ionic Liquid Having Property of Temperature-dependent Miscibility with Water; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, November 6-8, 2013, Tokyo Japan, B13-2.

K. Sasaki, T. Suzuki, T. Arai, K. Takao, Y. Ikeda: Electrochemical Studies on Uranyl Nitrate Complex in 1-Ethyl-3-methylimidazolium Nitrate; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, November 6-8, 2013, Tokyo Japan, B13-1.

T. Mori, K. Sasaki, T. Suzuki, T. Arai, K. Takao, Y. Ikeda: Separation of Uranyl Species Using Task-specific Ionic Liquid, [Hbet][Tf2N]; 2nd China-Japan Academic Symposium on Nuclear Fuel Cycle (ASNFC 2013), November 27-30, 2013, Shanghai, China, 7-6.

K. Takao, S. Takao, Y. Ikeda, G. Bernhard, C. Hennig: Uranyl-Halide Complexation in *N*,*N*-Dimethylformamide: Halide Coordination Trend Manifests Hardness of $[UO_2]^{2+}$; *33rd International Conference on Solution Chemistry (33ICSC)*, July 7-12, 2013, Kyoto, Japan, 4DO10.

Koichiro Takao: Facile and Universal Method for Ionic Liquid Preparation through Evaporation of HX Gas; *The 94th CSJ Annual Meeting*, March 27-30, 2014, Nagoya University, 4E4-09.

Koichiro Takao, Tomoya Suzuki, Masayuki Harada, Yasuhisa Ikeda: Consideration on Molecular Structure of Uranyl-Nitrate Precipitates Including Cycloalkylated 2-Pyrrolidone Derivatives; 2014 Annual Meeting of Atomic Energy Society of Japan, March 26-28, 2014, Tokyo Metropolitan University, G41.

K. Ploykrachang, H. Fukuda, K. Kondo, Y. Oguri, J. Hasegawa: Production of Quasimonochromatic X-Ray Microbeams Using MeV-Protons and a Polycapillary X-Ray Half Lens; *Int. J. PIXE*, Vol.23, No.1&2, pp.1-11 (2013).

Y. Oguri, Y. Hu, K. Kondo, H. Fukuda, J. Hasegawa: Digital Subtraction Cineangiography Using Proton-Induced Quasi-Monochromatic Pulsed X-Rays; *Int. J. PIXE*, Vol.23, No.1&2, pp.21-29 (2013). T. Kikuchi, Y. Oguri: Progress of High-Power Accelerator Research for Heavy-Ion Fusion - Final Focusing; *J. Plasma Fusion Res.*, Vol.**89**, No.2, pp.110-115 (2013).

Y. Oguri, K. Kondo, J. Hasegawa: Numerical Study of Heavy-Ion Stopping in Foam Targets with One-Dimensional Subcell-Scale Hydrodynamic Motions; *Nucl. Instrum. Meth. Phys. Res. Sect. A.*, Vol.**733**, pp.4-7 (2014).

J. Hasegawa, K. Kondo, Y. Oguri, K. Horioka: Ion Beam Focusing with Cone Optics for WDM Experiments; *Nucl. Instrum. Meth. Phys. Res. Sect. A.*, Vol.**733**, pp.32-38 (2014).

K. Kobayashi, Y. Kawamoto, W. El-Masry, M. Eto, H. Tokimura, T. Kaneko, Y. Obayashi, H. Mita, K. Kanda, S. Yoshida, H. Fukuda, Y. Oguri: Evolution of Interstellar Organics to Meteoritic and Cometary Organics: Approaches by Laboratory Simulations; *Proc. International Astrobiology Workshop 2013*, Sagamihara, Japan, November 28-30, 2013, pp.34-35.

Y. Oguri, K. Kondo, J. Hasegawa, K. Horioka: Numerical Analysis on Hydrodynamic Behavior of Ion-Beam-Heated Uranium Targets for Equation-of-State Studies under Extreme Conditions; to be published in *Proc. 4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, Japan, November 6-8, 2013.

T. Toriyama, S. Suzuki, K. Kawasaki, Y. Takagi, M. Nagai, H. Fukuda, K. Kondo, Y. Oguri: Effect of Vacuum Drying in Target Preparation from Potassium Hydroxide Solution on the Spectra of Rutherford Backscattering Spectroscopy; to be published in *Proc. 4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, Japan, November 6-8, 2013.

K. Kobayashi, Y. Kawamoto, M. Eto, T. Kaneko, Y. Obayashi, H. Mita, H. Fukuda, Y. Oguri, K. Kanda: Alteration of Complex Amino Acid Precursors by Soft X-Rays in Simulated Circumstellar and Interplanetary Environment; *13th European Workshop on Astrobiology (EANA 2013)*, Szczecin, Poland, July 22-25, 2013, MMP-4.

Y. Oguri, K. Kondo, J. Hasegawa, K. Horioka: Hydrodynamic Simulation for Equation-of-State Studies of High-Energy-Density Nuclear Materials Using High-Power Heavy-Ion Beams; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, Japan, September 3-5, 2013, F12.

Y. Oguri, Y. Hu, K. Kondo, J. Hasegawa: Proton-Induced Characteristic X-Ray Source for Deep-Seated Tumor Treatment Using a Syringe Needle; *11th European Conference on Accelerators in Applied Research and Technology (ECAART-11)*, Namur, Belgium, September 8-13, 2013, P99.

K. Ploykrachang, J. Hasegawa, K. Kondo, H. Fukuda, Y. Oguri: Development of a Micro-XRF System for Biological Samples Based on Proton-Induced Quasimonochromatic X-Rays; *11th European Conference on Accelerators in Applied Research and Technology (ECAART-11)*, Namur, Belgium, September 8-13, 2013, P101.

Y. Hu, K. Kondo H. Fukuda, K. Ploykrachang, Y. Oguri: Dose Distribution around a Syringe-Needle-Type Proton-Induced Quasi-Monochromatic X-Ray Source for Prostate Cancer Therapy; 2014 Annual Meeting of Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, D11.

M. Eto, H. Tokimura, R. Aratake, Y Kawamoto, T. Okabe, T. Kaneko, Y. Obayashi, S. Yoshida, H. Fukuda, Y. Oguri, K. Kobayashi: Formation of Complex Amino Acid Precursors and Nucleic Acid Bases by Particles Irradiation of Simulated Interstellar Media; *39th Annual Meeting of the Society for the Study of the Origin and Evolution of Life Japan*, Hiroshima, Japan, March 13-15, 2014, No. 20.

Rana Syed Masud, Hitoshi Mimura, Yuji Sasaki and Masaki Ozawa: Adsorption Properties of Rare Metals by Functional Xerogels Impregnated with MIDOA Extractant; 2013 Asian Pacific Conference on Chemical, Material and Metallurgical Engineering (APCCMME 2013), May 22-23, 2013, Beijing, China.

Masaki Ozawa: From Radioactive Wastes to Resources, by Neutronic Transmutation to Create New Rare Metals, Rare Earth; *PIM2013 Processes in Isotopes and Molecules*, Plenary Pl-11, Invited Lecture, 25-27 September 2013, Cluj-Napoca, Romania.

Masaki Ozawa, Chi Young Han, Toshitaka Kaneshiki, Masao Nomura, Shinichi Koyama, Mikael Nilsson: *Après ORIENT;* A P&T-based New Resource Strategy in Nuclear Fuel Cycle; *Global2013*, No.7477, 4x. Advanced Aqueous Separations, Salt Lake City, USA, September 29-October 3, 2013.

Yoshihiko SATO, Takehiro MATSNAGA, Shin-ichi KOYAMA, Tatsuya SUZUKI, Masaki OZAWA: Mechanism and Kinetics of thermal decomposition of tertiary pyridine resin in the nitrate form; *The fourth International Symposium on Innovative Nuclear Energy Systems -Innovative Nuclear Science and Technology Post Fukushiuima-INES-4*, 6th-8th November,2013, Tokyo Institute of Technology, Japan.

Masaki Ozawa: *Après ORIENT*, A New P&T Challenge to Transmute Radioactive Wastes into Resources; *The 2nd China-Japan Academic Symposium on Nuclear Fuel Cycle(ASNFC 2013)*, Abstract Book, pp64-65, Nov.27-Nov.30 2013, Shanghai, China. Atsunori Terashima, Masaki Ozawa: Numerical Analysis about Creation of Strategic Important Elements by Nuclear Transmutation Process of Fission Products; *The* 2nd China-Japan Academic Symposium on Nuclear Fuel Cycle(ASNFC 2013), Abstract Book, pp66-67, Nov.27-Nov.30 2013, Shanghai, China

Mikael Nilsson and Masaki Ozawa: Urban Mining of Used Nuclear Fuel as a Source of Valuable Materials; Symposium on New Frontiers in Sustainable Fuels and Chemicals "What's Beyond the Horizon", UC Santa Barbara, Thursday February 6, 2014.

Masaki Ozawa and Mikael Nilsson: Transmutation of Radioactive Waste into Resource, as "Kopernikanische Wendung"; 247th ACS National Meeting and Exposition, Industrial and Engineering Chemistry R&D in Processes for Waste Recycling and Metal Recovery "Symposium in Honor of Professor Jan Rydberg", March 18, 2014, Dallas, Texas.

Atsunori Terashima, Chi Young Han, Masaki Ozawa: Energy Dependence of the Neutron Transmutation from Spent Fuel to the Rare Earth Elements; *30th Symposium on Rare Earths*, Kitakyusyu, May 23-24, 2013, 1C-09.

Toshitaka Kaneshiki, Atsunori Terashima, Masao Nomura, Masaki Ozawa: Barium/Lanthanum Separation by Using Crown Ether Resins; *30th Symposium on Rare Earths*, Kitakyusyu, May 23-24, 2013, 1C-10.

Atsunori Terashima, Chi Young Han, Masaki Ozawa: Basic Research on the Element Transmutation of All the Fission Products Using Nuclear Reactors (1) Calculation of the Production Rate and Evaluation of Products; 2013 Fall Meeting of Atomic Energy Society of Japan, Hachinohe, September 3-5, 2013, N46.

Masaki Ozawa: Challenge of Nuclear Transmutation f or New Elements, Après ORIENT; *Rokkasho-mura Nu clear Fuel Cycle Seminar*, Oct.25, 2014.

A.Ohtaki, M.Hirata, Y.Sato, M.Ozawa: Element Strategy in Nuclear Fuel Cycle (III) – Resouceability of Nuclear Rare Earth; 2014 Annual Meeting of Atomic Energy Society of Japan, C02, Mar.26, 2014.

Byambajav Munkhbat, Toru Obara: Conceptual design of a small nuclear reactor for large-diameter NTD-Si using short PWR fuel assemblies, *Journal of Nuclear Science and Technology*; **50** [1], pp.46-58 (2013).

Masao Komeda, Kozo Kawasaki, Toru Obara: A new irradiation method with a neutron filter for silicon neutron transmutation doping at the Japan research reactor No.3 (JRR-3); *Applied Radiation and Isotopes*, **74**, pp.70-77, (2013).

Toru Obara, Taiki Onoe: Flattening of burnup reactivity in long-life prismatic HTGR by particle type burnable poisons; *Annlas of Nuclear Energy*, **57**, pp.216-220 (2013).

Dwi Irwanto, Toru Obara: Decay Heat Removal without Forced Cooling on a Small Simplified PBR with an Accumulative Fuel Loading Scheme; *Annlas of Nuclear Energy*, **60**, pp.383-395 (2013).

Piyatida Trinuruk, Toru Obara: Small, long-life high temperature gas-cooled reactor free from prompt supercritical accidents by particle-type burnable poisons; *Journal of Nuclear Science and Technology*, **50** (9), pp.898-912 (2013).

Piyatida Trinuruk, Toru Obara: Concept of prismatic high temperature gas-cooled reactor with SiC coating on graphite structures; *Annals of Nuclear Energy*, **63**, pp.437-445 (2013).

Toru Obara, Haruka Kikuchi: Energy Release in Criticality Accidents Involving Two Fuel Solution Tanks; *Transactions of the American Nuclear Society*, **109**, pp.888-889 (2013).

Topan Setiadipura, Toru Obara: Development of a Monte Carlo based PBR Fuel Management Code; *Transactions of the American Nuclear Society*, **109**, pp.1431-1433 (2013).

Piyatida Trinuruk, Toru Obara: Preventing the Prompt Critical Accident in Block-type HTGRs by using Particle Type Burnable Poisons; 2013 Fall Meeting of Atomic Energy Society of Japan, September 3-5, 2013, Hachinohe, Japan, G33 (2013).

M. Zamengo, J. Ryu, Y. Kato: Composite block of magnesium hydroxide-Eexpanded graphite for chemical heat storage and heat pump; *Applied Thermal Engineering*, **69**, pp. 29-38 (2014).

Odtsetseg Myagmarjav, Junichi Ryu, Yukitaka Kato: Lithium bromide-mediated reaction performance enhancement of a chemical heat-storage material for magnesium oxide/water chemical heat pumps; *Applied Tehrmal Eng.*, **63**, pp. 170-176 (2014).

Zamengo M., Ryu J., Kato Y.: Thermochemical performance of magnesium hydroxide - expanded graphite pellets for a packed bed reactor chemical heat pump; *Applied Thermal Engineering*, **64**(1-2), pp. 339-347 (2014)

Kato, Y.: Utilization of HTGR on active carbon recycling energy system; *Nuc. Eng. Design*, **271**, pp. 79-83 (2014).

Arnoldus Lambertus Dipu, Yutaka Ujisawa, Junichi Ryu, Yukitaka Kato: Carbon dioxide reduction in a tubular solid oxide electrolysis cell for acarbon recycling energy system; Nuc. Eng. Design, 271, pp. 30-35 (2014).

Seon Tae Kim, Junichi Ryu, Yukitaka Kato: The optimization of mixing ratio of expanded graphite mixed chemical heat storage material for magnesium oxide/water chemical heat pump; *Applied Therm. Eng.*, **66**, pp. 274-281 (2014).

Hiroyuki Sato, Xing L. Yan, Yukio Tachibana, Kazuhiko Kunitomi, Yukitaka Kato: Transient Analysis of Depressurized Loss-Of-Forcedcirculation Accident without Scram in High-Temperature Gas-Cooled Reactor; *Nuclear Technology*, **185**, pp.227-238, (2014).

Zamengo M., Ryu J., Kato Y.: Magnesium hydroxide expanded graphite composite pellets for a packed bed reactor chemical heat pump; *Applied Thermal Engineering*, **61**, 853-858 (2013).

Yukitaka Kato: (Invited Lecture) Innovative Low-Carbon Energy Sytem Based on HTGR; *CIGS Energy 2050 Symposium*, 24 Feb., 2014, Tokyo.

Yukitaka Kato: (Invited Lecture) Fture Vision of Japanese Energy System; *North-Kanto SCEJ Group*, 26 April, 2013, Gunma Univ.

Junichi RYU, Hirokazu ISHITOBI, Yukitaka KATO: Dehydration and Hydration Reactivity of La-based Mixed Hydroxides for Chemical Heat Storage; *International Sorption Heat Pump Conference (ISHPC 2014)*, March 31-April 3, 2014, Washington DC, USA.

Y. Hara, J. Ryu, Y. Kato: Kinetic performance study of Mg(OH)₂-Ni(OH)₂ composites prepared by mechanochemical method; *International Symposium on EcoTopia Science 2013*, Nagoya, 15 Dec., 2013.

Y. Kato: Active Carbon Recycling for Ironmaking and Future Energy Systems; *International Conference on Smart Carbon Saving and Recycling for Ironmaking (ICSRI)*, O-1, 2(2-4) October, 2013, Shonan Village Center, Japan.

Arnoldus Lambertus Dipu, Junichi Ryu, Yukitaka Kato: High-temperature Carbon Dioxide Electrolysis by Tubular SOEC for a Carbon Recycling Ironmaking Process; *International Conference on Smart Carbon Saving and Recycling for Ironmaking (ICSRI)*, O-10, 3 (2-4) October, 2013, Shonan Village Center, Japan.

M Zamengo, J Ryu, Y Kato: Volumetric Heat Storage Capacity in a Packed Bed Reactor of a Magnesium Oxide / Water Chemical Heat Pump; *International Symposium on Innovative Materials for Processes in Energy Systems* 2013 (*IMPRES2013*), Fukuoka, 4-6 Sep., 2013.

O Myagmarjav, J Ryu, Y Kato: Enhancement Of Reaction Rate Of Chemical Heat Storage Materials By Lithium Bromide For Chemical Heat Pump; International Symposium on Innovative Materials for Processes in Energy Systems 2013 (IMPRES2013), Fukuoka, 4-6 Sep., 2013.

Ishitobi, J Ryu, Y Kato: Kinetic Analysis of Hydration for Lithium Chloride-Modified Magnesium Oxide for Thermochemical Energy Storage; *International Symposium on Innovative Materials for Processes in Energy Systems 2013 (IMPRES2013)*, Fukuoka, 4-6 Sep., 2013.

Junichi RYU, Takafumi MIZUNO, Hirokazu ISHITOBI and Yukitaka KATO: Reactivity of Mg-Co Mixed Hydroxide as a Material For Chemical Heat Storage; *International Symposium on Innovative Materials for Processes in Energy Systems 2013 (IMPRES2013)*, Fukuoka, 4-6 Sep., 2013.

Kato: Contribution of thermal energy storage technologies on vehicle's thermal energy storage (VTES); *Massive Energy Storage for the Broader Use of Renewable Energy Sources*, Newport Beach, CA, USA, 23-26 June, 2013.

J Ryu, Ishitobi, Y Kato: Durability of chemically modified magnesium oxide for chemical heat storage; *Massive Energy Storage for the Broader Use of Renewable Energy Sources*, Newport Beach, CA, USA, 23-26 June, 2013.

M Zamengo, J Ryu, Y Kato: Magnesium hydroxide pellets for the storage of industrial waste heat", *Massive Energy Storage for the Broader Use of Renewable Energy Sources*, Newport Beach, CA, USA, 23-26 June, 2013.

O Myagmarjav, J Ryu, Y Kato: Reaction rate enhancement of chemical heat storage by lithium bromide for chemical heat pumps; *Massive Energy Storage for the Broader Use of Renewable Energy Sources*, Newport Beach, CA, USA, 23-26 June, 2013.

Y. Kato: Contribution of iACRES on Future; *ISIJ 166th Spring Meeting*, 22 March, 2014, Tokyo Tech.

G. Fujii, A. L. Dipu, J. Ryu, Y. Kato: CO₂ Decomposition by Solid Oxide Electrolysis Cell in Disk Type; *SCEJ* 79th *Annual Meeting*, 19 March, 2014, Gifu Univ.

H. Hatsuda, J. Ryu, Y. Kato: Metal Hydrogen Permeation Membrane Made by Reversed Build-Up Method; 19 March, 2014, Gifu Univ.

Odtsetseg Myagmarjav, Junichi Ryu And Yukitaka Kato: Mg(OH)₂-Expanded Graphite-LiBr composite (EML) for Middle-Temperature Thermal Energy Storage; *ISET'S 13*, Nagoya Univ., 13 Dec., 2013.

Yukitaka Kato, Reynold Dipu: Carbon Dioxide Electrolysis in Ni-YSZ|YSZ|LSM-YSZ Cell for a Carbon Recycling Iron-making System; *Regular Meeting of 54th*

Committee on ironmakin of JSPS, Tokyo, 29(28-29) Nov., 2013.

Y. Kato: Contirbution of thermochemical energy storage on low-carbon ironmaking system; *ISIJ 166th Autumn Meeting*, 17 Sep., 2013, Kanazawa Univ.

Y. Hara, J. Ryu, Y. Kato: Dehydraion/hydration reactivity of $Mg(OH)_2$ -Ni(OH)_2 complex thermochemical energy storage material prepared by mechano-chemical method; *SCEJ* 45th Autumn Meeting, 17 Sep., 2013, Okayama Univ.

Z. MASSIMILIANO, J. Ryu, Y. Kato: Temperature analysis in a packed bed reactor of a magnesium oxide-water chemical heat pump during hydration reaction; 50th HTSJ Symposium, 29 May, 2013, Sendai.

Jun Kariya: Thermal performance enhancement in a Calcium Oxide/Water chemical heat pump by employment of Expanded Graphite; *ANS 2013 Student Conference*, MIT, 6 (4 - 6) April, 2013, Boston, MA, USA (Poster).

T. Yamagishi, H. Nakano, Y. Kato: Heat storage apparaus; *Japan Patent 2013-246216*, submitted on 2 Dec., 2013.

Kazumi Kitayama and Hiroshige Kikura: Important Viewpoint on Disposal of Hi-level Radioactive Waste #3; *Denki-Jouhou, April*, pp. 32-46 (2013).

Keisuke Tsukada, Nobuyoshi Tsuzuki and Hiroshige Kikura: Ultrasonic Characterization on Air-coupled Ultrasound Flowmeter; *IEICE Technical Report*, Vol. **113**, No. 68, pp. 29-34 (2013).

Tomonori Ihara, Hiroshige Kikura and Yasushi Takeda: Ultrasonic Velocity Profiler for Very Low Velocity Field, *Flow Measurement and Instrumentation*, Vol. **34**, pp. 127-133 (2013).

Tat Thang Nguyen, Hiroshige Kikura, Ngoc Hai Duong, Hideki Murakawa and Nobuyoshi Tsuzuki: Measurements of Single-phase and Two-phase Flow in a Vertical Pipe using Ultrasonic Pulse Doppler Method and Ultrasonic Time-domain Cross-correlation Method; *Vietnam Journal of Mechanics*, Vol. **35**, No.3, pp. 239-256 (2013).

Thanh Tung Duong, Hiromasa Tanaka, Nobuyoshi Tsuzuki, Hideki Kawai and Hiroshige Kikura: An Investigation of Joule-heating flow in a cubic cavity using Finite Element method; *Nuclear Science and Technology*, Vol. **3**, No. 3, pp. 31-40 (2013).

Tat Thang Nguyen, Hideki Murakawa, Nobuyoshi Tsuzuki and Hiroshige Kikura: Development of Multiwave UVP and PIV Measurement Method to Measurements of Single-phase and Two-phase Bubbly Flow in a Vertical Pipe using Ultrasound; *PetroVietnam Journal*, Vol. **10**, pp. 36-47 (2013). Weerachon Treenuson, Nobuyoshi Tsuzuki, Hiroshige Kikura, Masanori Aritomi, Sanehiro Wada and Kenichi Tezuka: Accurate Flowrate Measurement on the Double Bent Pipe using Ultrasonic Velocity Profile Method; *Journal of Japanese Society for Experimental Mechanics*, Vol. **13**, No. 2, pp. 200-211 (2013).

Sanehiro Wada, Kenichi Tezuka, Weerachon Treenuson, Nobuyoshi Tsuzuki and Hiroshige Kikura: Estimating the Number of Transducers for Flow Rate Measurement using the UVP Method Downstream of Double Elbows; *Flow Measurement and Instrumentation*, Vol. **32**, pp. 51-62 (2013).

Tat Thang Nguyen, Ngoc Hai Duong and Hiroshige Kikura: Development and Application of Multiwave Method using Ultrasonic Pulse Doppler Method for Measuring Two-phase Flow; *Journal of Japan Society of Experimental Mechanics*, Vol. **13**, No.3, pp. 277-284 (2013).

Tat Thang Nguyen, Hiroshige Kikura, Ngoc Hai Duong and Hideki Murakawa: Study of Bubbly Two-phase Flows in a Vertical Pipe using Ultrasound and Image Processing; *Journal of Nuclear Science and Technology of Vietnam*, Vol. **3**, No. 3, pp. 13-24 (2013).

Yutaka Tamaura, Satoshi Shigeta, Quin-Long Meng, Taiki Aiba and Hiroshige Kikura: Cross linear solar concentration system for CSP and CPV; *Energy Procedia*, Vol. **49**, pp. 249-256 (2014).

Ari Hamdani, Nobuyoshi Tsuzuki, Daisuke Ito and Hiroshige Kikura: Two-Phase Observation of Taylor-Couette Vortex Flow Using Wire-Mesh Tomography; *Proceedings of the 8th International Conference on Multiphase Flow (ICMF2013)*, May, 2013, Jeju, South Korea, ICMF2013-548, pp. 1-5.

Thanh Tung Duong, Hiromasa Tanaka, Nobuyoshi Tsuzuki, Hideki Kawai and Hiroshige Kikura: Characteristics Observation of Joule-heating Flow Measurement Using UVP and PIV; *Proceedings of 8th International Conference on Multiphase Flow (ICMF2013)*, May, 2013, Jeju, South Korea, ICMF2013- 652, pp.1-5.

Takuya Fukumoto, Keisuke Tsukada, Tomonori Ihara, Nobuyoshi Tsuzuki and Hiroshige Kikura: A Study of Phased Array Ultrasonic Velocity Profile Monitor for Flow Rate Measurement; *The 21st International Conference on Nuclear Engineering (ICONE21)*, August, 2013, Chengdu, China, ICONE21-16601.

Tat Thang Nguyen, Hiroshige Kikura, Ngoc Hai Duong, Hideki Murakawa and Nobuyoshi Tsuzuki: Measurements of Bubbly Counter-current Two-phase Flow in a Vertical Pipe using Ultrasonic Doppler Method (UDM) and Ultrasonic Time Domain Cross-correlation (UTDC) Method; *Proceedings of the 14th Asia Congress of Fluid* *Mechanics - 14ACFM*, October, 2013, Hanoi and Halong, Vietnam, pp. 753-760.

Tat Thang Nguyen, Ngoc Hai Duong, Nobuyoshi Tsuzuki, Hideki Murakawa and Hiroshige Kikura: Measurements of Instantaneous Velocity Profiles of Single-phase and Two-phase Bubbly Flow in a Vertical Pipe using Ultrasound; *The 12th International Conference on Fluid Control, Measurement and Visualization (FLUCOME 2013)*, November, 2013, Nara, OS06-3-1.

Keisuke Tsukada, Nobuyoshi Tsuzuki and Hiroshige Kikura: Ultrasonic Characterization on Air-coupled Ultrasonic Flowmeter using Ultrasonic Beam Focusing; *The 12th International Conference on Fluid Control, Measurement and Visualization (FLUCOME 2013)*, November, 2013, Nara, OS14-4-1.

Ari Hamdani, Thang Tat Nguyen, Nobuyoshi Tsuzuki, Daisuke Ito and Hiroshige Kikura: The Application of Wire Mesh Sensor in Taylor Couette Flow; *The 4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, November, 2013, Tokyo, PNUCENE-D-13-00286, pp. 1-10.

Keisuke Tsukada, Nobuyoshi Tsuzuki and Hiroshige Kikura: A Study of Air-coupled Ultrasonic Flowmeter using Beam Focusing; *The 4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, November, 2013, Tokyo, PNUCENE-D-13-00304R2.

Thanh Tung Duong, Hiromasa Tanaka, Nobuyoshi Tsuzuki, Hideki Kawai and Hiroshige Kikura: Experimental and Numerical Investigation of Joule-heating Flow in Square Cavity; *The 4th COE-INES International Symposium on Innovative Nuclear Energy Systems (INES-4)*, November, 2013, Tokyo, C22-3.

Tomonori Ihara and Hiroshige Kikura: Optimization of the Ultrasound Buffer Rod for the Molten Glass Measurement; *The* 4th *COE-INES International Symposium on Innovative Nuclear Energy Systems* (*INES-4*), November, 2013, Tokyo, C23-2.

Weerachon Treenuson, Nobuyoshi Tsuzuki, Hiroshige Kikura and Sanehiro Wada: Out of Plane Effect on Double Bent Pipe Flow; *The International Conference on Experimental Mechanics 2013 (ICEM 2013) & the 12th Asian Conference on Experimental Mechanics (ACEM12)*, November, 2013, Bangkok, Thailand, EFO-124, 6 pages.

Thanh Tung Duong, Hiromasa Tanaka, Nobuyoshi Tsuzuki and Hiroshige Kikura: Numerical and Experimental Investigation on Chaotic flow in a Cubic Joule-heated Glass Melter; *Proceedings of 1st Vietnam-Japan Research of Human Resources Development Forum on Nuclear Technology*, December, 2013, Hanoi, Vietnam. Thanh Tung Duong, Hiromasa Tanaka, Nobuyoshi Tsuzuki, Hideki Kawai and Hiroshige Kikura: Effect of Cooling Condition on the Chaotic Flow Behavior in Joule-heating Cavity; *Proceedings of the 8th International Symposium on Measurement Techniques for Multiphase Flows*, December, 2013, Quangzhou, China, Sat36, pp. 1-5.

Keisuke Tsukada, Nobuyoshi Tsuzuki and Hiroshige Kikura: Ultrasonic Characterization on Air-coupled Ultrasound Flowmeter; *IEICE Technical Committee*, May, 2013, Tokyo, US2013-13.

Tat Thang Nguyen, Ngoc Hai Duong and Hiroshige Kikura: Application of UVP and PIV Measurement Methods to Measurements of Single-phase and Bubbly Two-phase Flows in a Vertical Pipe; *Proceedings of the Scientific and Technology Conference on The Intellect of PetroVietnam: Integration and Sustainable Development*, May, 2013, Hanoi, Vietnam, pp. 496-506.

Keisuke Tsukada, Tomonori Ihara and Hiroshige Kikura: A Study on Non-contacting Flowrate Measurement using Air-coupled Ultrasound; *Proceedings of 18th National Symposium on Power and Energy Symposium*, June, 2013, Chiba, C212, pp. 307-310.

Takuya Fukumoto, Keisuke Tsukada, Tomonori Ihara, Nobuyoshi Tsuzuki and Hiroshige Kikura: A Study of Phased Array Ultrasonic Velocity Profile Method for Flow Rate Measurement; *Proceedings of 18th National Symposium on Power and Energy Symposium*, June, 2013, Chiba, C213, pp. 311-314.

Antonin Povolny, Masaaki Tachi, Nobuyoshi Tsuzuki and Hiroshige Kikura: Flow Resistance Effect on Unstable Boiling Behavior in Parallel Channel Loop; *Proceedings* of 18th National Symposium on Power and Energy Symposium, June, 2013, Chiba, C222.

Hiromasa Tanaka, Thang Tung Duong, Nobuyoshi Tsuzuki and Hiroshige Kikura: Flow Behavior Measurement for Joule-heating Cavity with Cooled Electrodes by using UVP; *Proceedings of Symposium of the Visualization Society of Japan 2013*, July, 2013, Tokyo, D214.

Yutaka Tamaura, Nobuyoshi Tsuzuki and Hiroshige Kikura: Novel Cross Linear Solar Concentration System for CSP and CPV Solar Plants; 22nd Conference of Japan Institute of Energy, August, 2013, Tokyo, 4-5-1.

Thanh Tung Duong, Hiromasa Tanaka, Nobuyoshi Tsuzuki, Hideki Kawai and Hiroshige Kikura: An Investigation of Joule-heating flow in a cubic cavity using Finite Element method; *Proceedings of 10th Vietnamese National conference on Nuclear Science and technology*, August, 2013, Vung Tau, Vietnam, VINANST10-A11, pp. 1-10.

Ari Hamdani, Nobuyoshi Tsuzuki, Daisuke Ito and Hiroshige Kikura: Accuracy of Wire Mesh Sensor for Two-phase Flow Measurement in Taylor-Couette Flow; *Proceedings of Multiphase Flow Symposium of Japanese Society for Multiphase Flow (JSMF)*, August, 2013, Nagano, GS-3 B141, pp. 1-2.

Thanh Tung Duong, Hiromasa Tanaka, Nobuyoshi Tsuzuki, Hideki Kawai and Hiroshige Kikura: PIV Flow Measurement for chaotic flow induced by Joule-heating; *Proceedings of Multiphase Flow Symposium of Japanese Society for Multiphase Flow (JSMF)*, August, 2013, Nagano, GS-3 B142, pp. 1-2.

Hiromasa Tanaka, Thang Tung Duong, Nobuyoshi Tsuzuki and Hiroshige Kikura: Flow Behavior Measurement for Joule-heating Cavity with Changed Top Boundary Conditions; 2013 JSEM Annual Conference on Experimental Mechanics, August, 2013, Akita, C313.

Ari Hamdani, Nobuyoshi Tsuzuki, Daisuke Ito and Hiroshige Kikura: The Application of Wire Mesh Sensor in Taylor Couette Flow; *Proceedings of the 5th Multidisciplinary International Student Workshop* (*MISW2013*), August, 2013, Tokyo, 102, pp. 90.

Tat Thang Nguyen, Nobuyoshi Tsuzuki, Hideki Murakawa and Hiroshige Kikura: Measurements of Bubbly Counter-current Two-phase Flow in a Vertical Pipe using Ultrasound; *Proceedings of Mechanical Engineering Congress, 2013 Japan (MECJ-13) of JSME*, September, 2013, Okayama, J041011.

Tat Thang Nguyen, Nobuyoshi Tsuzuki and Hiroshige Kikura: Measurement of thermal-hydraulic parameters of bubbly flow in a vertical pipe using ultrasonic measurement and optical visualization; *Proceedings of Thermal Engineering Conference 2013*, September, 2013, Aomori, C144.

Tat Thang Nguyen, Hiroshige Kikura, Ngoc Hai Duong, Hideki Murakawa and Nobuyoshi Tsuzuki: Study of single-phase and bubbly two-phase flows in a vertical pipe using ultrasound and image processing; *Proceedings of the* 10th National Conference on Nuclear Science and Technology (Vietnam), October, 2013, Vũng Tàu, Vietnam, A12.

Tat Thang Nguyen, Hiroshige Kikura, Hideki Murakawa and Nobuyoshi Tsuzuki: Measurement of Bubbly Two-phase Pipe Flow using Multiwave Ultrasonic Pulsed Doppler Method and Wire Mesh Tomography; *Poster Short Presentation Session of INES-4&AESJ*, November, 2013, Tokyo, P-19.

Weerachon Treenuson, Nobuyoshi Tsuzuki and Hiroshige Kikura: Estimation of the Optimal Number of Ultrasound Transducers for Accurate Flowrate Downstream of Inclination Bent Pipe; *Poster Short Presentation Session/INES-4&AESJ*, November, 2013, Tokyo, A-18.

Hiromasa Tanaka, Thang Tung Duong, Nobuyoshi Tsuzuki and Hiroshige Kikura: Cooling Temperature Effect of Electrodes on Joule-heating Convection in a Cubic Cavity; *Poster Short Presentation Session of INES-4&AESJ*, November, 2013, Tokyo, A-23.

Hiroshige Kikura, Nobuyoshi Tsuzuki and Hiromasa Tanaka: A Fundamental Study on Joule-heating Convection in a Cubic Cavity; *National Congress on Magnetic Fluid*, November, 2013, Toyama.

Hiromasa Tanaka: A Fundamental Study on Ultrasonic Flow Profile Measurement in Joule-heating Convection; 7th Students' Research Workshop of the Atomic Energy Society of Japan Kanto-Koetsu Branch, February, 2014, Tokyo, A6.

Taiki Aiba, Hiroshige Kikura and Yutaka Tamaura: Development of Controlling System for Cross Linear Solar Reflector Using Image Processing; 53rd Students' Research Workshop of the Japan Society of Mechanical Engineers Kanto Branch, March, 2014, Tokyo, 1418.

Syeilendra Pramuditya, Minoru Takahashi: Core Design Study for Power Uprating of Integral Primary System PWR; *Annals of Nuclear Energy*, Vol. **59**, pp.16-24 (2013).

Asril Pramutadi Andi Mustaria, Minoru Takahashi: Metallurgical Analysis of Corroded Bellows of Bellow-sealed Valve in Lithium Flow System; *Fusion Engineering and Design*, Vol.**88**, Issue 4, pp.202-208 (2013).

T. Takeda, M. Stefanica: Study on Liquid Jet for Neutron Source of BNCT; *19th International Conference on Nuclear Engineering (ICONE20)*, October 24-25, 2012, Paris, France, ICONE20-45600.

Minoru Takahashi, Yun Gao, Marion Guihot, Asril Pramutadi: Overview of Studies on Lead-bismuth Technology in Tokyo Institute of Technology; *International Symposium on Nuclear Back-end Issues and the Role of Nuclear Transmutation Technology after the Accident of TEPCO's Fukushima Daiichi Nuclear Power Stations*, Kyoto, November 28, 2013.

Marion Guihot, Minoru Takahashi: Status of Studies and Future Subjects of Material Corrosion for Lead-bismuth Cooling System; *International Symposium on Nuclear Back-end Issues and the Role of nuclear Transmutation Technology after the accident of TEPCO's Fukushima Daiichi Nuclear Power Stations*, Kyoto, November 28, 2013.

Asril Pramutadi Andi Mustari, Minoru Takahashi: Corrosion of Cold Worked YUS409D Bellows of Bellow-sealed Valve in LFR and ADS Environment; 4th International Symposium on Innovative Nuclear Energy Systems (INES-4), Tokyo, November 6-8, 2013.

Tri Dan Le, Minoru Takahashi: Experiment of Heat Transfer Coefficient and Critical Heat Flux in Single Pin and Triangular Pin Array with Wire Wrap Spacer; *4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, November 6-8, 2013.

Yun Gao, Minoru Takahashi, Masao Nomura: Experimental Study on Diffusion of Ni in Lead-bismuth Eutectic (LBE); *4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, November 6-8, 2013.

Marion Guihot, Asril Pramudati, Minoru Takahashi: Corrosion and Material Strength Behavior of Welded Steel HCM12 in Liquid Lead-Bismuth Eutectic; *4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, November 6-8, 2013.

Noriaki Inaba, Minoru Takahashi: Exprerimental Study on Bubble Number Distribution with Bubble Condensation in Subcooled Flow Boiling; *4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, November 6-8, 2013.

Minoru Takahashi, Mihail Mihalache, Radu Secareanu, Alexandre Chapelat: Fundamental Study on Heavy Metal Freezing Behavior for Stop of Water Leakage from LWR Containment Vessel; *4th International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, November 6-8, 2013.

Minoru Takahashi, Toru Obara, Akio Minato, Shoji Uchida, Kenji Kikuchi, Masatoshi Kondo, Akito Nagata: Activities in CRINES for Generation IV International Forum, Lead-cooled Fast Reactor; *4th International Symposium on Innovative Nuclear Energy Systems* (*INES-4*), Tokyo, November 6-8, 2013.

Kenji Takeshita: Energy-related technologies "Nuclear Energy"; *Handbook of Chemistry*, Applied Chemistry, 7th ed., Maruzen, pp. 219-224 (2014).

Masahiko Nakase, Hitoshi Rokkaku, Kenji Takeshita: High-performance extraction operation using emulsion flow protected by surfactants in a liquid-liquid countercurrent centrifugal extractor; *Journal of Nuclear Science and Technology*, Vol. **50**, No. 7, pp. 723-730 (2013).

Masahiko Nakase, Hiroshi Kinuhata, Kenji Takeshita: Multi-staging for extraction of cesium from nitric acid by a single liquid-liquid countercurrent centrifugal extractor with Taylor vortices; *Journal of Nuclear Science and Technology*, Vol. **50**, No. 11, pp. 1089-1098 (2013).

Masahiko Nakase, Kenji Takeshita: Continuous back extraction operation by a single liquid-liquid centrifugal

extractor with Taylor vortices; *GLOBAL 2013 International Nuclear Fuel Cycle Conference*, September 29-October 3, 2013, Salt Lake City, Utah, USA, 7697, pp. 1017-1023.

Kenji Takeshita, Takeshi Ogata, Hiroshi Oaki, Yusuke Inaba, Atsunori Mori, Tsuyoshi Yaita, Shin-ichi Koyama: Extraction Chromatographic Separation of Am(III) and Eu(III) by TPEN-immobilized Gel; *GLOBAL 2013 International Nuclear Fuel Cycle Conference*, September 29-October 3, 2013, Salt Lake City, Utah, USA, 8200, pp. 554-558.

Kenji Takeshita, Hideharu Takahashi, Yasuhiro Jinbo, Akio Ishido: Restoration of Water Environment Polluted by Radioactive Cesium Released from Fukishima Daiichi NPP; *GLOBAL 2013 International Nuclear Fuel Cycle Conference*, September 29-October 3, 2013, Salt Lake City, Utah, USA, 8570, pp. 957-960. Kenji Takeshita: Restoration of Water Environment

Polluted by Radioactive Cesium Released from Fukushima Daiichi NPP; *The 9th Asia Pacific Conference on Sustainable Energy & Environmental Technologies* (APCSEET 2013), Narita, Japan, July 5-8, 2013, C15.

Masahiko Nakase, Kenji Takeshita: Numerical simulation and dynamic visualization of oil-water multiphase flow in various geometries of liquid-liquid centrifugal extractors; *The Japanese Society for Multiphase Flow JSMF Symposium on Multiphase Flow 2013*, Nagano, Japan, August 9-11, 2013, B215.

Masahiko Nakase, Kenji Takeshita, Yuji Sasaki: Effectof additional agents on multispecies mutual separation by a liquid-liquid countercurrent centrifugal extractor with Taylor vortices; *2013 Fall Meeting of the Atomic Energy Society of Japan*, Hachinohe, Japan, September 3-5, 2013, M15.

Hideharu Takahashi, Hiroshi Oaki, Yusuke Inaba, Kenji Takeshita: Development of Cesium Separation Technique by Hydrothermal Treatment using Subcritical Water; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, Japan, September 3-5, 2013, O39.

Yasuhisa Ikeda, Kenji Takeshita, Tamotsu Kozaki, Tsutomu Sato, Nobuaki Sato, Akira Kirishima, Hitoshi Mimura, Yuichi Nibori, Takayuki Sasaki, Tatsuya Suzuki, Yaohiro Inagaki, Kazuya Idemitsu: Basic Studies for Developing Rational Treatment and Disposal System of Radioactive Wastes Generated by Fukushima Dai-ichi Nuclear Accident (1) Outline of Research Plans and Topics; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, Japan, September 3-5, 2013, N24.

Kenji Takeshita, Yusuke Inaba, Hiroshi Oaki, Satoshi Uehara: Basic Studies for Developing Rational Treatment and Disposal System of Radioactive Wastes Generated by

Fukushima Dai-ichi Nuclear Accident (6) Development of H2-H2O Hydrogen-isotope Exchange Process for Removal of Tritium; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, Japan, September 3-5, 2013, N29.

Keita Watanabe, Tetsuji Yano, Naoko Ikezi, Tomohiro Amagasa, Tetsuo Kishi, Kenji Takeshita, Kazuhiro Minami, Eiji Ochi: Advanced Study on Vitrification Process (5) In-situ characterization of Glass Melting by Raman spectroscopy; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, Japan, September 3-5, 2013, P36.

Tatsuya Fukuda, Yoshio Nakano, Kenji Takeshita, Kazuhiro Minami, Eiji Ochi: Advanced Study on Vitrification Process (6) Thermal Decomposition of Major Nitrate in Simulated High-level Liquid Wastes; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, Japan, September 3-5, 2013, P37.

Masahiko Nakase, Mikiya Tanaka, Kenji Takeshita: Continuous mutual separation of lanthanides by a liquid-liquid countercurrent centrifugal extractor with Taylor vortices; *The 4th International Symposium on Innovative Nuclear Energy Systems*, Tokyo, Japan, November 6-8, 2013, B13-3.

Kenji Takeshita, Yusuke Inaba, Tsuyoshi Yaita, Shin-ichi Suzuki, Atsunori Mori: Separation of Am/Eu by Thermosensitive Gel with Multidentate Inclusion Compound TPEN -- Extraction Separation of Am/Eu and Extraction Mechanism --; *The 45rd Autumn Meeting of the Society of Chemical Engineers*, Nagoya, Japan, November 22-23, 2013, N123.

Kenji Takeshita, Hiroshi Oaki, Tsuyoshi Yaita, Shin-ichi Suzuki, Atsunori Mori: Separation of Am/Eu by Thermosensitive Gel with Multidentate Inclusion Compound TPEN -- Synthesis of Chromatographic Agent and Column Separation Tests --; *The 45rd Autumn Meeting of the Society of Chemical Engineers*, Nagoya, Japan, November 22-23, 2013, N124.

Yusuke Inaba, Kohei Otake, Kenji Takeshita: The Development of Heavy Metal Extraction Process by an Ultrasonic Wave; *The 45rd Autumn Meeting of the Society of Chemical Engineers*, Nagoya, Japan, November 22-23, 2013, N122.

Kenji Takeshita: Recent Progress of Clean-up Technology of Radioactive Fallout by Fukushima Daiichi Nuclear Power Plant Accident; *International Conference on Processes in Isotopes and Molecules (PIM 2013)*, Cluj-Napoca, Romania, September 25-27, 2013, P1-13.

Kenji Takeshita, Yusuke Inaba, Tsuyoshi Yaita, Shin-ichi Suzuki, Atsunori Mori: Extraction Chromatographic Separation of MA/Ln by Thermosensitive Gel immobilizing TPEN -- Separation Performance of MA/Ln and Extraction Mechanism --; *The 32nd Symposium on Solvent Extraction*, Nagoya, Japan, November 22-23, 2013, B-12.

Kenji Takeshita, Hiroshi Oaki, Tsuyoshi Yaita, Shin-ichi Suzuki, Atsunori Mori: Extraction Chromatographic Separation of MA/Ln by Thermosensitive Gel immobilizing TPEN -- Synthesis of chromatographic agent and column experiments --; *The 32nd Symposium on Solvent Extraction*, Nagoya, Japan, November 22-23, 2013, B-13.

Yusuke Inaba, Yongjie Li, Michal Cibula, Kenji Takeshita: Separation of palladium by novel hydrophobic thiodiglycolamide extractants; *The 32nd Symposium on Solvent Extraction*, Nagoya, Japan, November 22-23, 2013, B-09.

Kenji Takeshita, Yasuhiro Jinbo, Akira Ishido: Removal of Radioactive Cesium from Soil and Sewage Sludge contaminated by Fukushima Daiichi NPP Accident; *The 2nd China- Japan Academic Symposium on Nuclear Fuel Cycle (2nd ASNFC 2013)*, Shanghai, China, November 27-30, 2013, 3-6.

Tatsuya Fukuda, Yoshio Nakano, Kenji Takeshita, Kazuhiro Minami, Eiji Ochi: Thermal Decomposition Behavior of Nitrate Solution containing Di-*n*-butylephosphate in Vitrification Process; *The 2nd China- Japan Academic Symposium on Nuclear Fuel Cycle (2nd ASNFC 2013)*, Shanghai, China, November 27-30, 2013, 3-13.

Kenji Takeshita, Hiroshi Oaki, Tsuyoshi Yaita, Shin-ichi Suzuki, Atsunori Mori: Chromatographic separation of Am(III) and Eu(III) by porous silica coated with TPPEN-NIPA gel; 247th ACS National Meeting & Exposion, Dallas, USA, March 19, 2014, 115.

Yuto Matsuzawa, Masahiko Nakase, Kenji Takashita, Mikiya Tanaka: Effect of the geometries of fluid region in centrifugal extractor on multistage extraction (1) Oil-water dispersion and single component extraction performance; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, G23.

Masahiko Nakase, Yuto Matsuzawa, Kenji Takashita, Mikiya Tanaka: Effect of the geometries of fluid region in centrifugal extractor on multistage extraction (2) Multiple components separation performance; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, G24.

Akihiko Yamauchi, Yusuke Inaba, Kenji Takashita: Development of the Strontium Separation Process using Crown Ether-NIPA Gel; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, H04. Kenji Takashita, Hideharu Takahashi, Yusuke Inaba, Namiko Murayama: Removal of Cs from Hydrothermal Treatment Water by Novel Ferricyanide Absorbent; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, H06.

Kenji Takashita, Satoshi Uehara, Yusuke Inaba: Basic Studies for Developing Rational Treatment and Disposal System of Radioactive Wastes Generated by Fukushima Dai-ichi Nuclear Accident (15) Activity of Pt Catalyst under Polluted Water Conditions and Evaluation of Separation Process Scale; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, H18.

Keita Watanabe, Tetsuji Yano, Naoko Ikedi, Tomohiro Amagasa, Tetsuo Kishi, Kenji Takeshita, Kazuhiro Minami, Eiji Ochi: Advanced Study on Vitrification Process (7) Effect of composition of pseudo-HL waste solution on Cold Cap by in situ X-ray CT imaging; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, H43.

Tatsuya Fukuda, Yoshio Nakano, Kenji Takeshita, Kazuhiro Minami, Eiji Ochi: Advanced Study on Vitrification Process (8) Thermal Decomposition Behavior of Nitrate/DBP System in Cold Cap Formation; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, H44.

Kota Kawai, Tatsuya Fukuda, Yoshio Nakano, Kenji Takeshita, Kazuhiro Minami, Eiji Ochi: Advanced Study on Vitrification Process (9) Thermal Decomposition of Simulated High-level Liquid Wastes/Poroous Silica Glass mixtures in Temperature Rising Process; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, H45.

Imari Takatsuka, Tatsuya Fukuda, Keita Kanazawa, Kenji Takeshita: Cs Removal from Polluted Soil using Hydrothermal Treatment with Organic Reagent; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, Japan, March 26-28, 2014, I34.

Kyojiro Morikawa and Takehiko Tsukahara: Investigati on of Unique Protonic and Hydrodynamic Behavior of Aqueous Solutions Confined in Extended Nanospace s; *Israel Journal of Chemistry*, **54**, pp.1564-1572 (201 4).

Takehiko Tsukahara: ADVANCED SEPARATION AND DETECTION SYSTEM FOR RADIONUCLIDES USING MICROFLUIDIC DEVICE; *Radiation Interaction with Materials and Its Use in Technologies*, 65-66, (2014).

Ki Chul Park, Naokazu, Idota, Takehiko Tsukahara: Synthesis of NIPAAm-based polymer-grafted silica beads by surface-initiatedATRP using Me₄Cyclam ligands and the thermo-responsive behaviors for lanthanide(III) ions; *Reactive and Functional Polymers*, **79**, pp.36-46 (2014).

Makoto Uyama, Makoto Harada, Takehiko Tsukahara, Tetsuo Okada: Behavior of Polyhydric Alcohols at Ice/Liquid Interface; *Journal of Physical Chemistry* C, **117**, pp.24873 – 24882 (2013).

Takehiko Tsukahara, Hiroyasu Hotokezaka, Masayuki Harada, Yoshikuni Kikutani, Manabu Tokeshi, and Yasuhisa Ikeda: Highly Efficient Electrochemical Valence Control of Uranium in a Microfluidic Chip equipped with Microelectrodes; *Microfluid Nanofluid*, **14**, pp.989 - 994 (2013).

T. Tsukahara; Electrochemistry, vol. **82** (9), 777-781, (2014) [in Japanese].

K. Tani, K. Shinohara, T. Oikawa, H. Tsutsui, S. Miyamoto, Y. Kusama, T. Sugie: Effects of ELM mitigation coils on energetic particle confinement in ITER steady-state operation; *Nucl. Fusion* **52**, 013012 (2013).

S. Maeyama, A. Ishizawa, T.-H. Watanabe, N. Nakajima, S. Tsuji-Iio, H. Tsutsui: Numerical techniques for parallel dynamics in electromagnetic gyrokinetic Vlasov simulations; *Computer Physics Communications* **184**, 2462 {2473 (2013).

Yuuichi ASAHI, Akihiro ISHIZAWA, Tomo-Hiko WATANABE, Hiroaki TSUTSUI, Shunji TSUJI-IIO: Regulation of ETG turbulence by TEM driven zonal flows; 55th APS DPP Meeting of The American Physical Society, Nov 14-18, 2013, Denver, USA.

S. Hatakeyama, D. Shiojima, M. Miura, Z. Yao, H. Tsutsui, S. Tsuji-Iio, R. Shimada: Flywheel Generator System for Magnets of Small Tokamak Devices; 1PoCL-07, MT-23, Boston US (2013).

M. Sekine, S. Ikeda, N. Hayashizaki, T. Kanesue, M. Okamura: Multiple Species Beam Production on Laser Ion Source for electron beam ion source in Brookhaven National Laboratory; *Review of Scientific Instruments*, Vol.**85**, Issue 2, pp. 02B920-1/4(2014).

A. Yamaguchi, K. Sako, K. Sato, N. Hayashizaki and T. Hattori: Measurement of beam characteristics from C⁶⁺ laser ion source; *Review of Scientific Instruments*, Vol.**85**, Issue 2, pp. 02B921-1/3(2014).

Liang Lu, Toshiyuki Hattori, Noriyosu Hayashizaki, Takuya Ishibashi, Masahiro Okamura, Hirotsugu Kashiwagi, Takeshi Takeuchi, Hongwei Zhao, Yuan He: Development of high intensity linear accelerator for heavy ion inertial fusion driver; *Nuclear Instruments and Methods in Physics Research A*, Vol.**729**, pp. 133-137(2013).

D. Satoh, N. Hayashizaki, M. Yoshida: Development of

Better Quantum Efficiency and Long Lifetime Iridium Cerium Photocathode for High Charge electron RF Gun; *Proceedings of IPAC2013*, May 12-17, 2013, Shanghai, China, 327-329.

W. Zhou, N. Oshima, Z. Chen, K. Ito, B. E. O'Rourke, R. Kuroda, R. Suzuki, H. Yanagishita, T. Tsutsui, A. Uedono and N. Hayashizaki: Development of a sample chamber with humidity control for an atmospheric positron probe microanalyzer; *The 16th International Conference on Positron Annihilation (ICPA-16)*, August 19-24, 2012, University of Bristol, United Kingdom, Journal of Physics: ConferenceSeries 443 (2013) 012090-1/5.

Tooru Kobayashi, Kenichi Tanaka, Gerard Bengua, Noriyosu Hayashizaki, Tatsuya Katabuchi, Minoru Takahashi, Masanori Aritomi: Development of Liquid Lithium Target of ⁷Li(p,n)⁷Be Reactions for Accelerator Based BNCT Irradiation System; *World Congress on Medical Physics and Biomedical Engineering 2012*, May 26-31, 2012, Beijing, China, IFMBE Proceedings, 39 (2013) 1907-1910.

Noriyosu Hayashizaki, Nagayasu Oshima, Brian E. O'Rourke, Atsushi Kinomura: Development of induction buncher for high-efficiency pulsed positron beam system; 2014 Annual Meeting of the Atomic Energy Society of Japan, Tokyo, March 26-28, 2014, D05.

Naoki Yamano, Nakahiro Yasuda, Yoshinobu Izumi, Yoshihisa Matsumoto, Noriyosu Hayashizaki, Yoshihiko Sinoda: A Study on Nuclear Consensus Building (VI) - A study reviewing previous research on risk communication for low doses of ionizing radiation -; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, September 3-5, 2013, E39.

Kento Fumizawa, Noriyosu Hayashizaki, Tooru Kobayashi: Study on producing method of 98Mo(n, γ)99Mo with an accelerator-driven continuous neutron generator; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, September 3-5, 2013, F03.

Masaki Takamura, Noriyosu Hayashizaki: Design of a Liquidless Superconducting Cavity; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, September 3-5, 2013, F22.

Tatsunori Shibuya, Noriyosu Hayashizaki, Mitsuhiro Yoshida: Laser Ion Source for Generation of Highly Charged Ions and High Brightness; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, September 3-5, 2013, F24.

Megumi Sekine, Tatsunori Shibuya, Noriyosu Hayashizaki, Masahiro Okamura: Multiple Species Beam Production on Laser Ion Source; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, September 3-5, 2013, F25. Tomohiro Suzuki, Noriyosu Hayashizaki: Development of High Intensity Electron Gun with Thermionic Cathode; 2013 Fall Meeting of the Atomic Energy Society of Japan, Hachinohe, September 3-5, 2013, F29.

Sicheng Liu, Xunyue Liu, Radhika P. Kamdar, Rujira Wanotayan, Mukesh K. Sharma, Noritaka Adachi, Yoshihisa Matsumoto: C-terminal region of DNA ligase IV drives XRCC4/DNA ligase IV complex to chromatin; *Biochemical and Biophysical Research Communications.*, Vol. **439**, pp. 173-178 (2013).

Yoshihisa Matsumoto, Shoji Imamichi, Mikoto Fukuchi, Sicheng Liu, Rujira Wanotayan, Shingo Kuniyoshi, Kazuki Yoshida, Yasuhiro Mae, Mukesh K. Sharma: Radiosensitization strategy through modification of DNA double-strand break repair; *DNA Repair-New Research Directions*, Edit. Chen, C. ISBN 980-953-307-746-3, InTech (2013).

Yoshihisa Matsumoto: Biological Basis of Radiation Effects on Human Health; *Japanese Journal of Occupational Medicine and Traumatology*, Vol. **61**, pp. 180-185 (2013).

Mukesh K. Sharma, Radhika P. Kamdar, Shoji Imamichi, Mikoto Fukuchi, Sicheng Liu, Wanotayan Rujira, Yoshihisa Matsumoto: XRCC4: The genuine target of DNA-PK in non homologous end joining; 3rd Asian Congress of Radiation Research, Beijing (Beijing International Convention Center), China, 10-13 May 2013, Symposium (Oral) S9-5.

Mukesh K. Sharma, Shoji Imamichi, Yoshihisa Matsumoto: Detection of in cellulo phosphorylation of XRCC4 by DNA-PK; *The 15th Annual Meeting of the Biological Section of Japanese Society for Radiation Oncology*. Sendai (Tohoku University), 6 July 2013, Oral (3-4).

Shoji Imamichi, Yoshihisa Matsumoto, Mukesh K. Sharma: The effects of inhibitors for DNA-PK and ATM on the regulation of XRCC4 through phosphorylation in DNA double-strand break repair; *The 56th Annual Meeting of Japan Radiation Research Society*, Aomori (Crown Palace Aomori), 18-20 October 2013, Oral (O1-1-2).

Rujira Wanotyana, Mikoto Fukuchi, Mukesh K. Sharma, Yoshihisa Matsumoto: Role of XRCC4 C-terminal Part in DNA Double-Strand Break Repair Revealed by Systematically Generated Mutants; *The 56th Annual Meeting of Japan Radiation Research Society*, Aomori (Crown Palace Aomori), 18-20 October 2013, Oral (O1-6-5).

Mukesh K. Sharma, Shoji Imamichi, Radhika P. Kamdar, Mikoto Fukuchi, Sicheng Liu, Rujira Wanotayan, Yoshihisa Matsumoto: Identification of the phosphorylation sites of XRCC4 by DNA-PK essential for DNA double-strand break repair; *The 56th Annual Meeting* of Japan Radiation Research Society, Aomori (Crown Palace Aomori), 18-20 October 2013, Oral (O1-8-4).

Sicheng Liu, Rujira Wanotayan, Yoshihisa Matsumoto, Akinari Yokoya: Visualization of cell cycle arrest by X-irradiation and NHEJ inhibitors on Hela-Fucci cells; *The 56th Annual Meeting of Japan Radiation Research Society*, Aomori (Crown Palace Aomori), 18-20 October 2013, Oral (O1-12-3).

Yoshihisa Matsumoto: Tasks in radiation biology emerged after Fukushima Daiichi Nuclear Power Plant Accident; *The 56th Annual Meeting of Japan Radiation Research Society*, Aomori (Crown Palace Aomori), 18-20 October 2013, Oral (W9-2).

Yumi Sunatani, Radhika P. Kamdar, Mukesh K. Sharma, Osamu Matsui, Ryo Sakasai, Mitsumasa Hashimoto, Yoshihisa Matsumoto, Kuniyoshi Iwabuchi: Cleavage of XRCC4 by caspase and nuclear translocation of CAD; *The 56th Annual Meeting of Japan Radiation Research Society*, Aomori (Crown Palace Aomori), 18-20 October 2013, Oral (O1-8-1).

Yoshihisa Matsumoto: Mechanisms of the recognition and repair of DNA double-strand breaks and possible application in cancer therapy; *The Fourth International Symposium on Innovative Nuclear Energy Systems*, Tokyo (Tokyo Institute of Technology), Japan, 6-8 November 2013, Oral C32-1.

Tomohiro Kobayashi, Katsumi Yoshida, Toyohiko Yano: Effects of Heat-Treatment Temperature and Starting Composition on Morphology of Boron Carbide Particles Synthesized by Carbothermal Reduction; *Ceram. Int.*, Vol. **39**, pp.597-603 (2013).

Tomohiro Kobayashi, Katsumi Yoshida, Toyohiko Yano: Effects of Addition of Seed Grains on Morphology and Yield of Boron Carbide Powder Synthesized by Carbothermal Reduction; *Ceram. Int.*, Vol. **39**, pp. 3849-3856 (2013).

Noppasint Jiraborvornpongsa, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Effects of Trace Amount of Nanometric SiC Additives with Wire or Particle Shapes on the Mechanical and Thermal Properties of Alumina Matrix Composites; *J. Mater. Sci.*, Vol. **48**, No. 20, pp. 7022-7027 (2013).

Areerak Rueanngoen, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Recovery Behavior of Neutron Irradiated a- and b-SiAION Ceramics by Thermal Annealing up to 1473 K; *J. Nucl. Mater.*, Vol. **437**, No. 1-3, pp. 235-239 (2013).

Tomohiro Kobayashi, Katsumi Yoshida, Toyohiko Yano: Microstructure, Mechanical and Thermal Properties of B₄C/CNT Composites with Al Additive; *J. Nucl. Mater.*, Vol. 440, No. 1-3, pp. 524-529 (2013).

Katsumi Yoshida, Satoshi Kajikawa, Toyohiko Yano: Microstructure Design and Control for Improvement of Thermal Conductivity of SiC_f/SiC Composites; *J. Nucl. Mater.*, Vol. **440**, No. 1-3, pp. 539-545 (2013).

Toyohiko Yano, Yuusuke Futamura, Masamitsu Imai, Katsumi Yoshida: Recovery Behavior of Neutron-Induced Damage of AlN Irradiated at Higher Temperatures by Thermal Annealing; *J. Nucl. Mater.*, Vol. **440**, No. 1-3, pp. 495-499 (2013).

Yutaka Shinoda, Yoshikazu Suzuki, Katsumi Yoshida: TEM Analysis of Nanocrystalline SiC Ceramics Sintered by SPS Using Al₂O₃–TiO₂ Additive; *J. Asian Ceram. Soc.*, Vol. 1, No. 3, pp. 267-273 (2013).

Areerak Rueanngoen, Koumei Kanazawa, Masafumi Akiyoshi, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Effects of Neutron Irradiation on Polymorphs of Silicon Nitride and SiAION Ceramics; *J. Nucl. Mater.*, Vol. **442**, No. 1-3, Supplement 1, S394-S398 (2013).

Toyohiko Yano, Yusuke Futamura, Saishun Yamazaki, Takashi Sawabe, Katsumi Yoshida: Recovery Behavior of Point Defects after Low-Dose Neutron Irradiation at ~423 K of Sintered 6H–SiC by Lattice Parameter and Macroscopic Length Measurements; *J. Nucl. Mater.*, Vol. **442**, No. 1-3, Supplement 1, S399-S403 (2013).

Katsumi Yoshida: Chapter 18, Application of Electrophoretic Deposition for Interfacial Control of High-Performance SiC Fiber-Reinforced SiC Matrix (SiC_t/SiC) Composites; in *MAX Phases and Ultra-High Temperature Ceramics for Extreme Environments*, Eds. by I. M. Low, Y. Sakka, C. F. Hu, pp.533-552, IGI-Grobal, Hershey, PA, USA (2013).

Toyohiko Yano, Branko Matovic: Chap. 4.3, Advanced Ceramics for Nuclear Applications; in *Handbook of Advanced Ceramics*, 2nd Edition, Ed. by Shigeyuki Somiya, pp.353-368, Elsevier, ISBN: 9780123854698 (2013).

Branko Matovic, Toyohiko Yano: Chapter 3.1, Silicon Carbide and Other Carbides: From Stars to the Advanced Ceramics; in *Handbook of Advanced Ceramics*, 2nd *Edition*, Ed. by Shigeyuki Somiya, pp.225-244, Elsevier, ISBN: 9780123854698 (2013).

Noppasint Jiraborvornpongsa, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Synthesis Mechanism of SiC Nanowires Fabricated by Thermal Evaporation Method; *Seventh International Conference on Science and Technology of Advanced Ceramics (STAC-7)*, June 20, 2013, Yokohama, Japan, 2E-05.

Tomohiro Kobayashi, Katsumi Yoshida, Toyohiko Yano: Effect of Sintering Additives on Mechanical Properties of B₄C/CNT Composites; Seventh International Conference on Science and Technology of Advanced Ceramics (STAC-7), Yokohama, Japan, June 20, 2013, PP-032.

Toyohiko Yano, Yan You, Koumei Kanazawa, Masamitsu Imai, Katsumi Yoshida, Saishun Yamazaki: Recovery Behavior of Neutron-Irradiation-Induced Point Deffects of High-Purity b-SiC; 16th International Conference on Fusion Reactor Materials (ICFRM16), Beijing, China, October 23, 2013, 16-452, A093.

Areerak Rueanngoen, Koumei Kanazawa, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Analysis of Recovery Process of Low Dose Neutron Irradiation-Induced Defects in Silicon Nitride-Based Ceramics by Thermal Annealing; *16th International Conference on Fusion Reactor Materials (ICFRM16)*, Beijing, China, October 23, 2013, 16-540, A110.

Katsumi Yoshida, Hiroyuki Akimoto, Akihiro Yamauchi, Toyohiko Yano, Masaki Kotani, Toshio Ogasawara: Interface Formation of Unidirectional SiC_f/SiC Composites by Electrophoretic Deposition Method; 2nd International Symposium on Inorganic and Environmental Materials (ISIEM2013), Rennes, France, October 28, 2013, B1-1-1.

Noppasint Jiraborvornpongsa, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Synthesis and Growth Mechanism of SiC/SiO₂ Core-Shell Nanowires by Thermal Evaporation Method; *International Symposium on Inorganic and Environmental Materials (ISIEM2013)*, Rennes, France, October 28, 2013, 8.

Katsumi Yoshida, Hiroyuki Akimoto, Toyohiko Yano, Masaki Kotani, Toshio Ogasawara: Mechanical Properties of Unidirectional and Crossply SiC_f/SiC Composites Using SiC Fibers with Carbon Interphase Formed by Electrophoretic Deposition Process; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, Japan, November 7, 2013, B23-2.

Areerak Rueanngoen, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Microstructure of Low-Dose Neutron Irradiated Silicon Nitride-Based Ceramics after Thermal Annealing; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, Japan, November 7, 2013, B23-4.

Hiroshi Konishi, Katsumi Yoshida, Masamitsu Imai, Toyohiko Yano: Neutron Irradiation Effect of Oxide Sintering Additives for SiC_f/SiC Composites; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, Japan, November 8, 2013, P-18.

Mohd Idzat Idris, Hiroshi Konishi, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Neutron Irradiation Swelling of SiC and SiC_f/SiC for Advanced Nuclear

Applications; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, Japan, November 8, 2013, P-21.

Masafumi Akiyoshi, Toshimasa Yoshiie, Xu Qiu, Koichi Sato, Toyohiko Yano: Annealing Behavior of Thermal Diffusivity in Ceramics Irradiated by Electron and Neutron; *The Fourth International Symposium on Innovative Nuclear Energy Systems (INES-4)*, Tokyo, Japan, November 7, 2013, B23-3.

Noriyuki Watanabe, Masanori Aritomi and Toyohiko Yano: Geometrical Network Approach for an Evaluation of Heat-Transfer Coefficient in Dropwise Condensation for Passive Safety Systems; *The Fourth International Symposium on Innovative Nuclear Energy Systems* (*INES-4*), Tokyo, Japan November 7, 2013, C22-1.

Takuya Aoki, Toshio Ogasawara, Yosuke Okubo, Katsumi Yoshida, Toyohiko Yano: Fabrication of SiC/SiC Composites by Low Temperature Melt Infiltration; *The 13th JAPAN International SAMPE Symposium & Exhibition (JISSE-13)*, Aichi, Japan, November 12, 2013.

Katsumi Yoshida: Development of High Performance SiC_f/SiC Composites Based on Electrophoretic Deposition Process; 2nd Conference of The Serbian Ceramic Society, Belgrade, Serbia, June 6, 2013, PL-2.

Katsumi Yoshida: Interphase Coatings for High Performance SiC_f/SiC Composites by Electrophoretic Deposition Method; δ^{th} International Conference on High Temperature Ceramic Matrix Composites (HT-CMC8), Xi'an, China, September 25, 2013, HT-CMC8-S5-0064.

Katsumi Yoshida, Hiroyuki Akimoto, Akihiro Yamauchi, Toyohiko Yano, Masaki Kotani, Toshio Ogasawara: Application of Electrophoretic Deposition Method for Interphase Formation in SiC Fiber-Reinforced SiC Matrix Composites; *Korean Ceramic Society 2013 Fall Meeting*, Jeju, Korea, October 18, 2013, CS2-11.

Katsumi Yoshida: Development of Novel Interphase Formation Process for SiC_f/SiC Composites by Electrophoretic Deposition Method; 4th Seminar of R&D Center for High Reliable-High Temperature Materials, 2013, Tokyo, Japan, October 22, 2013.

Go Kametani, Takenobu Sakai, Shuichi Wakayama, Katsumi Yoshida: Evaluation of Crack Growth Resistance of Silicon Nitride under Thermal Shock Fracture by Acoustic Emission Technique; 19th Annual Meeting of The Japan Society of Mechanical Engineers, Kanto Branch, Tokyo, March 16, 2013, 20312, pp.277-278.

Noppasint Jiraborvornpongsa, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano, Tohru S. Suzuki, Yoshio Sakka: Effects of Strong Magnetic Field and Sintering Methods on the Microstructure of Al₂O₃/SiC Nanowires Composites; Annual Meeting of The Ceramic Society of Japan, 2013, Tokyo, March 17, 2013, 1P003.

Tomohiro Kobayashi, Katsumi Yoshida, Toyohiko Yano: Mechanical and Thermal Properties of Boron Carbide/Carbon Nanotubes Composites; *Annual Meeting of The Ceramic Society of Japan, 2013*, Tokyo, March 19, 2013, 3H06.

Areerak Rueanngoen, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Thermal Annealing Behavior of Neutron Irradiated-Si₃N₄ and SiAlON Ceramics; *Annual Meeting of The Ceramic Society of Japan, 2013*, Tokyo, March 19, 2013, 3H22.

Yosuke Okubo, Toyihoko Yano, Katsumi Yoshida, Toshio Ogasawara, Takuya Aoki: Fabrication of SiC_f/SiC Composite Fabricated by Advanced Melt Infiltration Method; 26th Fall Meeting of The Ceramic Society of Japan, Nagano, September 5, 2013, 2119.

Katsumi Yoshida, Hiryuki Akimoto, Akihiro Yamauchi, Yuto Hattori, Toyohiko Yano, Masaki Kotani, Toshio Ogasawara: Formation of Carbon- and BN-Interphases for SiC_f/SiC Composites by EPD Method and Their Mechanical Properties; 26^{th} Fall Meeting of The Ceramic Society of Japan, Nagano, September 5, 2013, 2120.

Tomohiro Kobayashi, Katsumi Yoshida, Toyihiko Yano: Effects of CNT Content and Sintering Additives on Thermal and Mechanical Properties of $B_4C/CNTs$ Composites; 26^{th} Fall Meeting of The Ceramic Society of Japan, Nagano, September 5, 2013, 2PI03.

Noppasint Jiraborvornpongsa, Masamitsu Imai, Katsumi Yoshida, Toyohiko Yano: Effects of Starting Materials on the Formation of SiC Nanowires by Thermal Evaporation Method; 26th Fall Meeting of The Ceramic Society of Japan, Nagano, September 5, 2013, 2PI04.

Katsumi Yoshida, Chin-Chet See, Satoshi Yokoyama, Toyohiko Yano: Effect of Sintering Temperature and SiC Particle Size on the Microstructure of Porous SiC Ceramics with In-Situ Grain Growth; 127th Academic Conference of the Society of Inorganic Materials, Japan, Yonezawa, November 15, 2013, (54), pp.108-109.

H. Sagara, H. Tomikawa, M. Watahiki and Y. Kuno: Feasibility study of passive gamma spectrometry of molten core material from Fukushima Dai-ichi nuclear power plant unit 1, 2 and 3 cores for special nuclear material accountancy - Low-volatile FP & special nuclear material inventory analysis and fundamental characteristics of gamma-ray from fuel debris -; *J. Nucl. Sci Technol*, vol. **51**, no. **1**, pp. 1-23 (2014).

K. Ismailov, K Nishihara, M. Saito, H. Sagara: Optimization study on accelerator driven system design for effective transmutation of Iodine-129; *Ann. Nucl. Energy*, vol. **56**, pp.136-142 (2013). E. Hamase, F. Damian, C. Poinot-salanon, M. Saito, H. Sagara, C. Y. Han: A feasibility study on long-life reduced-moderation water reactor with highly protected Pu breeding by doping with minor actinides; *Ann. Nucl. Energy*, vol. **54**, pp.76-84 (2013).

H. Sagara, T. Shiba, H. Tomikawa, A. Ishimi, K. Hori, and M. Saito: Passive Gamma Spectrometry of Low-Volatile FPs for Accountancy of Special Nuclear Material in Molten Core Material of Fukushima Daiichi Nuclear Power Plant -Numerical Simulation of Leakage Gamma-ray from Molten Core Materials in Hypothetical Canister-; *Trans. Am. Nucl. Soc.*, Vol. **108**, pp. 2316-2317 (2013).

C. G. Bathke, N. Inoue, B. B. Ebbinghaus, Y. Kuno, T. Mihara, H. Sagara, J. Murphy, D. Dalton, Y. Nagayama: Summary of a Joint US-Japan Study of Potential Approaches to Reduce the Attractiveness of Various Nuclear Materials for Use in a Nuclear Explosive Device by a Terrorist Group; *Proc. GLOBAL 2013*, pp. 230-236 (2013).

Hiroshi Sagara, Hirofumi Tomikawa, Akihiro Ishimi, Masaru Watahiki, and Yusuke Kuno: Passive Gamma Spectrometry of Low-Volatile FPs for Accountancy of Special Nuclear Material in Molten Core Material of Fukushima Daiichi Nuclear Power Plant -Overview and Research Plan; *Proc. INMM 54th mtg.* (2013).

Hiroshi Sagara, Koji Matsumoto, Ippei Yamauchi, Chi Young Han, Takashi Onishi, Shin-ichi Koyama, Masaki Saito: Feasibility study of the fast breeder reactor blanket as a target of plutonium denaturing and TRU transmutation for enhancement of proliferation resistance and actinide management in the future fuel cycle; *INES-4*, Tokyo, Nov. 6 - 8, 2013.

Tomooki Shiba, Hiroshi Sagara, Chi Young Han, Masaki Saito: Blanket Fuel with High Proliferation Resistance in Fast Breeder Reactor (XV) Prediction of irradiation behavior of DU-Am pellets; *2013 Fall Meeting of Atomic Energy Society of Japan*, Aomori, September 3-5, 2013, L26.

Kenji Nakao, Hiroshi Sagara, Masaki Saito: Material Balance and Proliferation Resistance of Multi-Fuel-Cycle with Fast Breeder Reactors; 2014 Annual Meeting of Atomic Energy Society of Japan, Tokyo, March 26-28, 2013, K12..

K. Kondo, J. Hasegawa, K. Horioka, Y. Ogur: Velocity measurement of electromagnetically-driven shock wave to produce a dissociated hydrogen target for beam interaction experiment; to be published in Proc. 4th International Symposium on Innovative Nuclear Energy Systems (INES-4), Tokyo, Japan, November 6-8, 2013.

K. Kondo, T. Moriyama J. Hasegawa, K. Horioka, Y.

Oguri: Electro-magnetically driven shock and dissociated hydrogen target for stopping power measurement; Nuclear Instruments and Methods in Physics Research A, Vol. **733**, pp.1-3 (2014).

K. Kondo, Y. Oguri: Velocity measurement of electro-magnetically-driven shock wave for dissociated hydrogen target; 2013 Autumn Meeting of Physical Society of Japan, September 25-28, 2013, Tokushima, Japan, 27aKC-6.

S. Fujioka, Z. Zhang, K. Ishihara, T. Ikenouchi, T. Johzaki, A. Sunahara, N. Yamamoto, H. Nakashima, K. Kondo, T. Watanabe, H. Sakagami, J. J. Santos, G. Lorenzo, Y. Arikawa, H. Nagatomo, K. Mima, H. Nishimura, H. Azechi: Application of laser-produced strong magnetic field to fast-ignition laser fusion, 30th Annual Meeting of The Japan Society of Plasma Science and Nuclear Fusion Research, December 3-6, 2013, Tokyo, Japan, 04aB04.

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