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### Abbreviation of the "BULLETIN OF THE LABORATORY FOR ADVANCED NUCLEAR ENERGY" is BULL. LAB. ADV. NUCL. ENERGY

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#### BULLETIN OF THE LABORATORY FOR ADVANCED NUCLEAR ENERGY BULL. LAB. ADV. NUCL. ENERGY, Vol.4, 2019

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I. Special Article in Commemoration of Retiring Professor

### I.1

### **Environmental Behaviors of Radionuclides**

Toshihiko OHNUKI

#### 1. Introduction

The presence of actinides in radioactive wastes is a major environmental concern due to their long radioactive half-lives, their high energy radiation emissions, and their chemical toxicity. In order to estimate the mobility of actinides from the waste disposal sites several studies examined its interactions with soils and subsoils composed of abiotic and biotic components, principally minerals and bacteria. Among the biotic components, several microorganisms have been shown to sorb actinides. The high capacity of microbial surfaces to bind actinides may affect the migration of actinides in the environment. However, we have only limited knowledge of the role of microorganisms in the migration of actinides in the environment.

In many geological disposal concepts, some kind of clay-based backfill composed of clays, is embedded between waste materials and geological system. One of the important barrier properties is high actinides sorption capacity to retard the migration of actinides. At the boundary of backfill material and geological system, clays are released and penetrated into fracture in geological system. Since some microorganisms are present in the fracture, actinides released from the repository migrated through mixture of clay and microorganisms. However, the effects of microorganisms on the sorption of actinides on the mixture of microorganisms and clay is not fully understand.

The severe accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) occurred when a tsunami struck along the east coast of Honshu, Japan, as a result of a huge earthquake (magnitude 9.0) that occurred on 11 March 2011. FDNPP eventually lost the ability to cool the nuclear fuel, which resulted in a series of hydrogen explosions [1, 2]. The explosions occurred from 13 to 15 March at Units 1 and 3, causing release of radioactive materials into the atmosphere and subsequent dispersal over the entire Northern Hemisphere [3]. These radioactive materials were then deposited on the ground not only in Fukushima Prefecture, where FDNPP is located [4, 5], but also in neighboring prefectures [6] and the adjacent Pacific Ocean [7]. The spatial concentration distribution and depth profiles of radioactive Cs in soils were measured [8-11] to estimate the dose rate and to estimate the fate of radionuclides in the terrestrial environment.

Radioactive particulates, termed Cs microparticles (CsMPs), were discovered in air dust samples collected from 9 pm on 14 March to 9 am on 15 March 2011 at Tsukuba, ~170 km south of FDNPP [12]. The analyses by radiochemical techniques and energy-dispersive X-ray (EDX) spectrometry indicated a high content of not only radioactivity but also high concentrations of radioactive Cs in the CsMPs. Consequently, many researchers have analyzed CsMPs, and these studies have revealed that the elemental composition consisted of Si, Fe, Zn, and Cs as major components, and K, Cl, Sn, Rb, Pb, and Mo as minor components. Many CsMPs were shown to be present 20 km northwest [19] and 3 km south [14] of FDNPP. The presence of CsMPs was also confirmed from analysis of dust on non-woven fabric cloth and needles of Japanese cedar [15, 16]. These findings revealed that CsMPs were widely dispersed not only in Fukushima, but in other more distant areas.

The CsMPs have occasionally been found to be associated with trace concentrations of U [14, 17, 18]. Ochiai et al. [17] reported that the CsMPs contained nanoscale fuel fragments in the form of  $UO_{2+X}$  nanocrystals and isometric (U,Zr) $O_{2+X}$  nanocrystals. Recently, the Tokyo Electric Power Corporation (TEPCO) reported that the fuel debris fragments collected in and around the primary containment vessels (PCVs) at FDNPP contained  $UO_{2+X}$  and (U,Zr) $O_{2+X}$  [19].

In this paper, the author summarizes our findings on the interactions of uranium [20] and Pu [21], with microorganisms under aerobic condition. In addition, the author also summarizes Characteristics of the CsMPs containing not only radioactive Cs, but also U and other heavy elements.

# 2. Association of actinides with microorganisms and clay

In this section, we summarize our findings on the interactions of uranium, one of the actinides, with

microorganisms in an aerobic condition. Uranium has two different oxidation states of U(VI) and U(IV). Hexavalent uranyl aqueous complexes is more mobile than U(IV) in an aerobic environment. Accumulation experiments of U(VI) on a mixture of microorganism and kaolinite clay were carried out. The association of U(VI) with the mixture and the yeast are directly analyzed by transmission electron microscopy (TEM). The effects of exudates on the sorption of of Eu(III) and Cm(III) on *Chlorella vulgaris* and cellulose which is major components of cell of *C. vulgaris* were examined [22].

# 2.1. Adsorption of U(VI) by a mixture of Bacillus subtilis and kaolinite clay

The accumulation of U by mixtures of *Bacillus* subtilis and kaolinite were investigated [20]. *B. subtilis* and kaolinite were chosen in the experiments even though smectite is major component clay in backfill material. Because (a) both are ubiquitous in the terrestrial environment; and, (b) their surfaces are well characterized.

A mixture of Bacillus subtilis (IAM 1069) and commercial kaolinite from Nihon Chikagaku-sha Co. Ltd., Kyoto, Japan were contacted with a  $4x10^{-4}$  M U(VI) solution for 48 h at pH 4.7±0.1 in an aerobic condition. No U(VI) was precipitated in a mixture-free 4×10<sup>-4</sup> M U(VI) solution at pHs between 3 and 5 for 96 h, indicating that U(VI) was undersaturated compared to U(VI)-oxy/hydroxides. Forty-eight hours after exposure to U solutions, desorption of U from the mixtures were carried out by a 1 M CH<sub>3</sub>COOK solution at pH 4.7 for 8 h. The oxidation states of U accumulated on the mixture were analyzed by X-ray absorption near edge structure (XANES). The mixture adsorbing U was observed by TEM (JEOL JEM-2000FXII), operated at 200 kV under standard conditions. Unstained samples were prepared by drying diluted (×200) aliquots of suspension on holey carbon films (a whole mount).

Our results showed that approximately 10% of U was removed by kaolinite, and about 40% by B. subtilis. Removal of uranium increased to approximately 30% with increasing the fraction of B. subtilis up to 5% in the mixtures. These results indicated that the mixtures of microorganism and kaolinite accumulate U(VI). We also showed the evidence of no precipitation of uranium on the surface of B. subtilis and kaolinite in the mixture by TEM and EDS analyses. No uraninite formation is consistent with the XANES analysis that oxidation state of U is VI (Fig. 1). These results suggest that U precipitation is not a dominant accumulation mechanism in the mixtures of B. subtilis and kaolinite under the experimental conditions. Thus, adsorption of U(VI) on the surface of B. subtilis and kaolinite is the dominant mechanism in the accumulation of U(VI) by the mixtures.

Approximately 80% of U was desorbed by the CH<sub>3</sub>COOK from the sample containing kaolinite alone, while about 65% was removed from the sample containing 95% kaolinite and 5% *B. subtilis* and from the sample containing *B. subtilis* alone. TEM and EDS analysis of the U accumulated mixture (Fig. 2) indicated that the EDS

spectrum of *B. subtilis* cells (Fig. 2b) showed a distinct peak of U M  $\Box$  and small peaks of Al K  $\Box$ , Si K  $\Box$ , P K  $\Box$ , and S K  $\Box$ . By contrast the EDS spectrum of the kaolinite particles revealed distinct peaks of Al K  $\Box$  and Si K  $\Box$  whereas  $\Box$  no peak of U M  $\Box$  was detected. These results verified that almost all of the U was associated with the cells of *B. subtilis* in the mixture.



Fig. 1 X-ray absorption near edge structure of U adsorbed on the mixture of *B. Subtilis* and kaolinite [20].



Fig. 2 (a) TEM photograph of the mixture and (b) EDS spectra of *B. subtilis* and kaolinite [20].

Kelly et al. [23] showed that U(VI) is associated with both protonated phosphoryl and deprotonated carboxyl groups on the cell surface of B. subtilis at pHs between 1.7 and 4.8. This association is in good agreement with the surface complex model study by Fowle et al. (2000) for *B*. subtilis. Kohler et al. [24] pointed out that U(VI) is mainly associated with the aluminol group of kaolinite. These results suggest that U(VI) forms surface complexes with the functional groups of the cellular surface of B. subtilis and/or edge sites of kaolinite. Approximately 80% of the accumulated U was desorbed with the CH<sub>3</sub>COOK solution from kaolinite, a result implying that the U bound by the silanol and aluminol groups is desorbable with CH<sub>3</sub>COOK. We assume that the U(VI) complex with the carboxyl group should have nearly the same stability constant as that with acetic acid because of the structural similarity of these functional groups. Accordingly this supposition leads to the idea that the U(VI) associated with carboxyl functional groups could be desorbed with CH<sub>3</sub>COOK. Therefore, the remaining fraction of U(VI) after desorption from B. subtilis with CH3COOK might be bound strongly to structures other than the carboxyl functional groups, i.e., phosphoryl groups on the surface of B. subtilis.

#### 2.2. Effects of exudates on Kd of Eu(III) and Cm(III)

In the experiments, adsorption kinetics and the distribution coefficients ( $K_d$ ) of Eu(III) and Cm(III) were

determined as function of solution pH. The concentrations of Eu and Cm in the suspension containing the algae were approximately  $1 \times 10^{-6}$  M and  $1 \times 10^{-8}$  M, respectively. Radionuclides of  $^{152}$ Eu and  $^{244}$ Cm were added for the measurement of the concentrations. The distribution coefficient, K<sub>d</sub> (ml/g), was calculated by Eq. (1)

Κ

$$_{\rm d} = (C_0 - C) V/CW \tag{1}$$

where  $C_0$  is initial concentration of metal ion in the solution; C the equilibrium concentration of the metal in the solution; V the volume of solution (ml); and W is the weight of the C. vulgaris on a dry weight. Adsorption of Eu(III) and Cm(III) on cellulose was measured using cellulose powder obtained from Kanto Chemical, Tokyo, Japan. The cellulose was used without purification.

The amounts of organic carbon exudated from *C. vulgaris* cells during incubation, possibly including the constituents of dead and dying cells, were determined as dissolved organic carbon (DOC). Approximately 0.025 mg of the algal cells was added to 5 ml of distilled water. The concentration of DOC in the filtrate was determined by total organic carbon analyzer, TOC-5000 (Shimadzu, Kyoto, Japan). The distribution coefficients of Eu and Cm for cellulose were obtained in the exudates solution that was collected from *C. vulgaris*.

Fractions of Eu and Cm sorbed on C. vulgaris as a function of time at pH 4, 5, and 6 (Fig. 3) showed that the sorbed fractions increased immediately after contact the elements with *C. vulgaris* cells. The maximum sorbed fractions were attained within 3 min, when all values exceeded 75%. Both elements showed higher sorbed fractions in the lower pH solution. Beyond 3 min the sorbed fractions decreased with increasing exposure time. No marked difference in sorption was observed between Eu and Cm.

Figure 4 shows the log  $K_d$  of Eu and Cm for *C. vulgaris* and cellulose in a 0.5% NaCl solution of pH between 2 and 9. For *C. vulgaris* log  $K_d$  of Eu and Cm rose with an increase in pH from 2 to 3 by a unit of 1; beyond pH 3, the log  $K_d$  declined with increasing pH. No significant difference was observed between log  $K_d$  of Eu and Cm within the pH range examined. The highest log  $K_d$  was approximately 4.0, observed at pH 3; the lowest one was 2.1 at pH 8. Adsorption of Eu and Cm on cellulose was almost identical. The log  $K_d$  of Eu and Cm increased with increasing pH from 2 to 6. Log  $K_d$  at pH 2 and pH 6 were approximately 0.7 and 5.1, respectively. Beyond pH 6, log  $K_d$  decreased with increasing pH, and it was 4.0 at pH 9.

The amount of DOC excreted from *C. vulgaris* cells showed a slight increase wih an increase in pH. This result indicated that exudates were released from the cells during the sorption experiments. The log  $K_d$  of Eu and Cm for cellulose in the exudates solution were plotted in Fig. 4. The log  $K_d$  of Eu and Cm were almost identical and both showed a tendency to decrease with an increase in pH with values of approximately 2.3, 1.9, and 1.7 at pH 4, 5, and 6, respectively.

The pH dependence of Eu and Cm sorption on cellulose differed from the one observed for *C. vulgaris*. The log  $K_d$  versus pH plots from pH 2 to 6 showed a positive slope.

The slope of log  $K_d$  for *C. vulgaris* showed a negative slope at pH between 3 and 8. On the contrary, the log  $K_d$  for cellulose in the exudates solution showed a negative slope at pH between 4 and 6. These results indicated that the exudates changed sorption behavior of Eu and Cm on cellulose, and resembled to that on *C. vulgaris*.



Fig. 3 Time course of adsorption of Eu(III) and Cm(III) by Chlorella vulgaris [22]



Fig. 4 Measured logKd of Eu(III) and Cm(III) for *C. vulgaris* and cellulose in the solution of different pH. The logKd of Eu(III) and Cm(III) for cellulose in the solution containing wxudartes from *C. vulgaris*.

Our results strongly suggested that exudates released from bacterial cells affect sorption behavior of actinides on bacterial cells.

# 2.3. Effect of microorganisms on the migration of actinides in backfill materials

High radiation dose and high temperature prevent activity of microorganisms in buffer materials in HLW repository. Although some microorganisms can survive under such extreme conditions, population of microorganisms may be not as high as in host rock. These suggest that microorganisms not directly affect the migration of radionuclides in buffer materials. On the other hand, microorganisms are survived in host rock outside of buffer materials.

It is well known that a small fraction of bentonite in buffer materials are leached out from repository, and diffused into the fracture of host rock (Alonso, 2007), where microorganisms are present. These bentonite colloids adsorb radionuclides, and migrate into the host rock (Nagasaki, 1999; Kurosawa, 2001). When the radionuclides adsorbed bentonite are leached from repository to the neighboring host rock, radionuclides are encountered to bentonite and microorganisms. Our results suggest that radionuclides, probably U and other actinides prefer to be accumulated in microorganisms in the mixture of microorganisms and kaolinite. Kaolinite is 1:1 clay mineral and smectite which is major clay mineral in bentonite is 2:1 clay mineral. Adsorption of radionulides on edge sites of these clays are nearly the same each other. Interlayer of smectite provide reversible adsorption site for cationic radionuclides except for Cs [25]. These facts suggest that some portion of radionuclides adsorbed on bentonite colloids are dissociated from bentonite colloids and are associated with microorganisms. It is well known that some microorganisms are present in groundwater (planktonic) and on rock (biomat). When microorganisms form biomat, the radionuclides may be eliminated from groundwater and migration of radionuclides is retarded by the association with biomat. On the contrary, if planktonic some microorganisms are populated, radionulides associated with the planktonic microorganisms are still present in groundwater. The radionuclides associated with the planktonic microorganisms are transported to far field by transportation and by diffusion. Thus, culture condition of microorganisms is very important to estimate colloidal migration of radionuclides in near field.

Microorganisms cultured in host rock around backfill materials excrete the exudates as described above. Dissolved organic substances in groundwater or buffer porewater may form complexes with radionuclides, thereby affecting their migration behavior. In backfill material, organic substances with large molecular size are expected to be retarded by the filtration effect of compacted bentonite [26]. The filtration and retardation of large molecular weight organic substances has been confirmed by infiltration experiments using polyacrylic acid [27]. These results indicate that only low molecular weight organic substances are capable of affecting the solubility, diffusion and sorption of radioelements in the buffer material [28]. Size of exudates from Saccharomyces cerevisiae were measured by size exclusive column to be approximately several tens molecular weight. This size is smaller than fulvic and humic substances, suggesting the exudates can diffuse into backfill material. Since microbial activity is not extensive in backfill materials, the exudates diffused in backfill materials are not degraded by microorganisms. These suggest that exudates are formed complexes with actinides in buffer materials, and enhance migration of actinides.

# **3.** Formation of radioactive cesium microparticles originating from the Fukushima Daiichi Nuclear Power Plant accident

In this section, the author introduces the characteristics of the CsMPs, and highlights the fact that the fuel debris fragments contained nanoparticles with the U and Zr components having similar structures to that of the CsMPs. This similarity in structures has stimulated further research on the structure and elemental constituents, especially for U and Zr in the CsMPs, and has opened up new avenues for studying the chemical characteristics of the fuel debris.

#### 3.1. Characteristics of CsMPs

The nanostructure and chemical properties of the CsMPs have been studied using scanning transmission electron microscopy energy-dispersive X-ray analysis (STEM-EDX) [14, 17, 18]. We focused on measurement of the CsMPs dispersed toward the western zone, which were

not derived from Unit 1. The elemental map of the focused ion beam (FIB)-prepared specimen of the CsMPs [14] showed that the distribution of the major elements appeared to be homogenous. The particle matrix was amorphous as revealed by the diffuse diffraction maxima in selected-area electron diffraction (SAED) patterns. Similarly, a uniform distribution of Si in amorphous material was shown by the transmission electron microscopy (TEM) analyses.

Uranium was detected in one of the CsMPs by microcalorimetric EDX analysis [18]. The presence of U in CsCl inclusion-sized particles of less than 20 nm diameter (**Fig. 5**) was confirmed by atomic-resolution electron microscopic analysis for CsMPs collected near FDNPP [14]. Uranium was present in amorphous form and only franklinite was found to be associated with the small amount of U.



Fig. 5 HAADF-STEM image of the closed area of the CsMP showing aggregation of franklinite nanoparticles (elemental peaks of Fe, Zn, and Si in the EDX spectrum [right upper]). Comparison of EDX spectra (edx1 and 2 in red lines) of the point analysis with the spectrum obtained by the area analysis (black line) revealing the presence of U peaks [14].

The thermal history of the CsMPs is relevant to understanding the conditions that led to the formation of the CsMPs as a result of the accident. Furuki et al. identified that the mineral franklinite contained W, besides U; W has a very high melting point of 3687 K, but in the oxidized form melts at ~1746 K [29]. It is known that the oxidation of U oxide fuel can decrease the volatilization temperature of U from 2,700 K to ~1,900 K in the case of the non-oxidized form of tetravalent uranium oxide (UO<sub>2</sub>) [30]. These facts suggest volatilization of the slightly oxidized form of UO<sub>2</sub> in the fuel occurred as a result of the accident. In addition, fuel fragments were incorporated into the Fe oxide nanoparticles before the molten core concrete interaction (MCCI).

Fragments of nuclear fuel bearing U were discovered [17]. In the CsMPs samples collected at Ottozawa located  $\sim 4$  km west of FDNPP, Fe oxide nanoparticles of  $\sim 100$  nm size were attached to the CsMP surface (indicated by the white square in **Figure 6**A). Two U oxide nanoparticles of

~10 and ~30 nm in size were present (see Fig. 6B) The Fe oxide and U oxide nanoparticles were identified as magnetite (Fe<sub>3</sub>O<sub>4</sub>) and uraninite (UO<sub>2+X</sub>), respectively (Figure 6C), with epitaxial growth between magnetite and uraninite being evident (Fig. 6C). For a portion of the U oxide nanoparticle, the high-resolution high-angle annular dark field (HR-HAADF)-STEM image (Figure 6C) displayed an enhanced contrast every 4 or 5 atomic columns, both in the a and b axes (Figure 6D). This was due to the overlying or underlying Fe atoms being in the same orientation along the projected zone axis because magnetite and uraninite are both isometric with unit cell parameters of 0.83958 [31] and 0.54682 nm [32], respectively.



Fig. 6 TEM characterization of the uraninite fragments in CsMP collected at Ottozawa in Okuma, Fukushima Prefecture [17]. (A) HAADF-STEM image of half of the CsMP and the STEM-EDX map of Cs La, of which the color represents the X-ray counts. (B) HAADF-STEM image of the same Fe-U particle observed in the CsMP after thinning by FIB and the elemental maps of selected elements. The white square and line segment AB are further characterized in panels C and G, respectively. (C) Magnified image of the area indicated by the white square in panel B and the SAED pattern, which were collected from the area including both Fe oxide and U oxide nanoparticles. Indexes in white and yellow letters correspond to magnetite and uraninite structures, respectively. The white square is further magnified in panel D. (D) HR-HAADF-STEM image of the area indicated by the white square in panel C. Fe atoms (red) of magnetite structure and U atoms (green) of uraninite structure are overlaid in the image. An atomic-column elemental map of U is also included. (E) Electron tomography showing the three-dimensional structure of uraninite nanoparticles in a magnetite nanoparticle. See the movie in the Supporting Information of Reference [17] for further details. (F) Cross-sectional view of uraninite nanoparticles embedded with the magnetite nanoparticle. A part of the euhedral rectangular shape can be observed, although the top and bottom of this magnetite nanoparticle was removed by the FIB thinning. (G) STEM-EDX line scan analysis of the traverse AB in panel B.

Ochiai et al. undertook three-dimensional electron

tomography of the magnetite nanoparticle, and this revealed the presence of two euhedral uraninite nanocrystals completely enclosed in a part of the euhedral magnetite crystal and oriented in the same direction, which is consistent with the crystallographic relation of epitaxial growth (Figure 6E and 6F). The magnetite nanoparticle was also associated with trace concentrations of Tc and Mo at the exterior (Figure 6G). Hence, the magnetite containing the uraninite nanoparticles was formed and impurities were not detected by EDX (detection limit ~0.1 wt %). Technetium was volatilized in the reactors and subsequently was adsorbed on the surface of magnetite. The presence of Tc indicated that the uraninite nanocrystals were embedded in magnetite nanocrystals which originated from inside the reactors. Based on this evidence, the nanoscale fragments originated from the reactor debris.



Fig. 7 TEM characterization of the uraninite fragments in CsMP collected at an aquaculture center ~2 km south of FDNPP[17]. (A) HAADF-STEM image of CsMP and the elemental maps of Fe and Cs. (B) Enlarged image of the area indicated by the white square in panel A associated with the elemental maps of Fe and U.

Other types of uraninite were also detected. For instance, uraninite nanoparticles were embedded in Fe oxide encapsulated in CsMPs (**Fig. 7A**). The texture of the CsMPs was indicative of aggregation of individual dense CsMPs. Within the porous portion of the CsMPs, a euhedral Fe oxide nanocrystal of ~400 nm size was found, which also contained an embedded U dioxide nanoparticle of ~70 nm in size (**Figure 7B**). This U-dioxide nanoparticle had a similar morphology, composition, and host phase as those shown in **Fig. 6**. The Fe oxide nanoparticle was embedded within the CsMP, indicating that the Fe oxides nanoparticles were encapsulated during the CsMP formation in the reactors, rather than becoming attached during dispersal in the atmosphere. Given that the CsMPs

were formed inside the reactors, the Fe oxide nanoparticles with the embedded U oxide nanoparticles originated from within the reactors as well.

The formation of U-Zr oxide nanoparticles was also demonstrated by their presence in the CsMPs collected at Ottozawa in Okuma, Fukushima Prefecture. The HAADF-STEM image of aggregates of the CsMPs [17] showed more than three spherical CsMPs were in an aggregated form. The presence of not only Cs but also U and Zr was confirmed by elemental mapping. The Zr and U distributions were almost homogeneous within the individual grains. The molar fractions of U in the U-Zr oxides, U/(U+Zr), determined by STEM-EDX analysis, varied widely between the particles, ranging from 0.14 to 0.91 (Fig. 9). A further thinning of the sample resulted in the detection of uraninite and cubic zirconia, revealing that these particles were single crystals and that one particle had pores of ~6 nm diameter and a few defects. The U/(U+Zr) molar ratios were 0.91 for one particle and 0.17 for another particle, indicating that U-Zr oxide solid solutions had been formed.



Fig. 9 Molar ratios of U/(U+Zr) analyzed using STEM-EDX [17]. The ratios reveal the average values: n = 7 for particle 1, n = 5 for particle 2, n = 2 for particles 3, 5, 6, and 7, and n = 1 for the others without error bar. The error bars show the standard deviations ( $2\sigma$ ).

# 3.2 Perspectives on CsMP analyses for understanding the characteristics of the fuel debris.

Analyses of the CsMPs dispersed as a result of the FDNPP accident have been carried out by different techniques. Detailed analyses by TEM and SR-X-ray have clarified the structure of the CsMPs and confirmed the presence of many elements including U and Fe, besides radioactive Cs. The U nanoparticles and the Fe oxide nanoparticles observed in the samples collected at Okuma had uraninite (UO<sub>2+X</sub>), X < 0.33, and magnetite structures, respectively. The origin of Fe in the magnetite was likely to

have been the stainless steel of the PCV and/or alloys present in the PCV.

The chemical form of Fe in the fuel debris at FDNPP was considered to be metallic based on the accident analysis calculations [40, 41]. On the other hand, magnetite particles were present in the CsMPs [17, 18]. Iron was also present in the fuel debris fragments collected from the PCVs [19]. The chemical forms of Fe in the fuel debris fragments have not yet been clarified. The chemical form of Fe may change during vaporization; that is, the vaporized Fe can easily bond to oxygen to form Fe oxide nanoparticles, mainly magnetite [42]. This vaporization results from partial vaporization of molten Fe metal above the melting temperature of 1811 K [36].

Takano et al. showed the effects of  $B_4C$  on the chemical forms of the fuel debris [43]. Iron containing borate materials were formed after melting of the various core materials [B<sub>4</sub>C, stainless steel, Zr, ZrO<sub>2</sub>, (U,Zr)O<sub>2</sub>] by arc melting. These results indicated that chemical forms of the Fe-containing fragments deposited on the fuel debris resulted from the melting and deposition processes of the fuel rods and the materials in the PCVs. Thus, the chemical forms of Fe-containing fragments deposited on the fuel debris should be examined in the future.

The presence of U in the CsMPs may aid in the clarification of the properties of the fuel debris deposited inside and outside of the PCVs. The wide range in the composition of U-Zr oxides (U/(U+Zr) = 0.14-0.91) indicates that the eutectic mixture of U-Zr oxides occurred heterogeneously at the microscale due to spatial variations in the chemistry. The presence of cubic U-Zr oxide nanoparticles with composition similar to the ZrO<sub>2</sub> end-member in the CsMPs suggest a more rapid cooling process than the one that occurred at Unit 2 of the Three Mile Island Nuclear Generating Station, where U-Zr oxide was reported to be tetragonal. The facts regarding the presence of U-rich (U,Zr)O2 cubic forms and Zr-rich (Zr,U)O<sub>2</sub> tetragonal forms in the fuel debris fragments of Unit 1 sediment (Fig. 9) are consistent with microscale heterogeneity existing during the formation of the (U,Zr) oxides.

A part of the process that the FDNPP fuels experienced during the event can be summarized as follows [17]: i) After the loss of power to the cooling system, the coolant water vaporized and the steam reacted with Zr and Fe resulting in the formation of the respective oxides.

ii)  $UO_2$ , the main component of the fuels, partially oxidized and then volatilized at temperatures greater than ~1900 K.

iii) The fuel assemblies melted unevenly with relatively less-irradiated fuels being heated to a higher temperature as compared with the high burnup fuels and then volatilized as evidenced by the  $^{235}$ U/ $^{238}$ U isotopic ratio values.

iv) The fuel assembly collapsed and moved to the bottom of the reactor pressure vessel. The temperature increased locally to at least greater than 2400 K based on the liquidus temperature of U–Zr oxides.

v) Locally formed oxides melted yielding a heterogeneous composition, including a small amount of Fe oxides, which then became a source of Fe–U single crystals and U–Zr

oxide eutectic phases. Specifically, euhedral magnetite nanocrystals encapsulated euhedral uraninite nanocrystals, which would have crystallized slowly at this stage.

vi) Liquid U–Zr oxide nanodroplets were rapidly cooled and solidified to a cubic structure. When the molten fuels hit the concrete pedestal of the PCV, SiO gas was generated at the interfaces between the melted core and concrete and instantly condensed to form CsMPs.

vii) The U–Zr oxide nanoparticles or the magnetite nanocrystals subsequently formed aggregates with the CsMPs. Finally, the reactor debris fragments were released into the environment along with the CsMPs.

Uraninite nanocrystals without detectable impurities were embedded within the Fe oxide. This scenario is similar to that illustrated in **Fig. 10** [19], where the cubic form of  $UO_2$  was present along with Fe-containing material and amorphous SiO<sub>2</sub>. These analyses indicated the presence of  $UO_2$  as particles around the PCVs of Units 1, 2, and 3. In addition, U was present with Fe-containing material, not only in the sediment material, but also in the particles collected on top of the PCVs. The Unit 2 particles were collected from the operating floor located at the top of the Unit 2 building, indicating that the particles were generated as a result of the meltdown. Interestingly, the Cs content in the particles was below the detection limit by EDX. This result strongly suggests that "hot" particles containing U but not radioactive Cs were present near the reactor.



Fig. 11 Elemental mapping of U and Fe by SEM-EDX analyses of the particles collected from inside the PCV of Unit 3 in July 2017 [19]. The particles may have originated from the sediments around the pedestal.

Thus, further research not only on the CsMPs but also on the particles around the reactors of Units 1, 2, and 3 should be pursued to better understand the characteristics of the fuel debris and to further clarify the processes involved in the FDNPP accident.

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# II. Research Reports

A. Innovative Nuclear Energy System Division

# A.1 Study on Innovative Nuclear Reactor Designs and Technology for Fukushima Daiichi NPS Decommissioning

### Toru OBARA

Studies on innovative nuclear reactor designs and Technology for Fukushima Daiichi Nuclear Power Station (NPS) have been performed. The studies were focused on the Breed and Burn reactors, High Temperature Gas-cooled reactors, neutron flux measurement in high gamma-ray background, and criticality safety in fuel debris retrieval.

# 1. Study on stationary wave breed and burn reactor concept

In this study, a rotational fuel shuffling concept was proposed. In this concept, fuel assemblies are moved to the next position step by step in a divided symmetry core region. Fresh fuel is loaded from the periphery and moved toward the center region, then moved outward and discharged. If the core could achieve an equilibrium state at which high reactivity fuels are continuously placed in the core center region; it would be possible to retain the breed and burn regions stationary. In such kind of equilibrium placed in high state, high reactivity fuels are neutron-importance region stably. Α small lead-bismuth-cooled fast reactor with metallic fuel was adopted as the core design [1].

# 2. Study on small CANDLE burning reactor with melt-refining process

Maintaining the fuel cladding integrity of a CANDLE reactor during operation is one of the key technological challenges that still need to be addressed. The introduction of the melt-refining process has shown great potential for solving this challenge in the high-burnup condition of large CANDLE reactors with metal fuel. The purpose of the study was to design a small CANDLE reactor with the melt-refining process. The analysis focused on the impacts of core homogenization, melt-refining, and the cooling and waiting times of the fuel. From the equilibrium analysis, the burning region velocity, neutron flux distribution, power density distribution, and core nuclide number density distribution were obtained [2].

# 3. Study on CANDLE burning fast reactor using plutonium from LWR spent fuel

Analyses were performed to determine the initial core design of a CANDLE burning fast reactor with a minimum amount of plutonium from LWR spent fuel. The initial core was designed with a starter fuel to initiate the reactor, and a booster fuel to increase the effective multiplication factor to more than unity. This reactor was able to maintain criticality with an excess reactivity of less than 1% throughout the operating period. The temperature coefficient was calculated, while it could be negative at the beginning of the operation, it was positive thereafter. Thermal hydraulic analyses were also performed. A fuel could be cooled to less than a criteria temperature at the equilibrium state, but during the transient period, the fuel could not be cooled even with the maximum coolant inlet velocity of 2.0 m/s. In order to make it possible to cool fuel to less than the criteria, reactor power was decreased so that the maximum temperature was lower than the limitation temperature [3].

# 4. Study on MOX and Pu-ROX fuels in small pebble bed reactor with accumulative fuel loading scheme

The accumulative fuel loading scheme was introduced as the simplest fueling strategies for a Pebble Bed Reactor (PBR). In this study, mixed-oxide (MOX) and plutonium-rock like oxide (Pu-ROX) fuels were introduced as starting materials. The optimum fuel composition of MOX fuel was 5g HM/pebble when uranium-plutonium ratio was 70:30. In the case of MOX fuel, the maximum discharged burnup was 172 GWd/t over 6 years of operation time. On the other hand, the optimum fuel composition for Pu-ROX fuel was 3g HM/pebble. The results showed that the maximum discharged burnup was 550 GWd/t with an operation time of 12.7 years. A negative temperature coefficient was achieved for both MOX and Pu-ROX fuels throughout the operation period [4].

# 5. Study on Supercritical transient analysis in fuel-debris systems

The purpose of this study was to confirm the applicability of the multi-region approach to supercritical transient in hypothetical fuel-debris systems with more complicated geometry and composition than those of our previous studies. This study also aimed to investigate the relative impact of several fuel-debris parameters on the supercritical transient. A total of fourteen hypothetical systems were made by varying the three considered parameters. As a result of this study, a region-wise power profile, temperature profile, and energy release were obtained for each system. For the systems studied herein the parameter with the largest impact on the supercritical transient was found to be the enrichment of <sup>235</sup>U, followed in most cases by the angle of repose of the conical fragmented debris region, while the height of the consolidated debris region generally had the least impact. It was shown that the multi-region approach can be a useful tool for analysis of supercritical transient in fuel-debris systems with rather complicated geometry and composition [5].

6. Study on liquid activation circulation material to measure the neutron flux in a high gamma-ray

#### background

The activation detector is the most convenient, especially against a high gamma-ray background. In this study, liquid activation material is introduced to obtain a continuous measurement, and a rhodium solution is used to measure the reaction rate. There are three regions in this concept: the activation region, transfer or circulation region, and measurement or detection region. In this study,  $Rh_2(SO_4)_3$  with the concentration of 100 g/L is used as the solution. To obtain the maximum reaction rate, an optimum ow rate of 80 cm<sup>3</sup>/s was applied. Results showed a maximum counting rate in the equilibrium condition of about 18.6cps. It is possible to use a liquid activation method to measure the neutron flux in a high gamma-ray background [6].

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### A.2 Li<sub>4</sub>SiO<sub>4</sub>/CO<sub>2</sub> Chemical Heat Pump for High Temperature Gas Cooled Reactors

#### Yukitaka KATO, Seon Tae KIM and Hiroki TAKASU

#### **1. INTRODUCTION**

Nuclear energy is an excellent source of process heat for various industrial applications and the only credible non carbon option. Especially, high temperature gas cooled reactors (HTGRs) which has the capacity to provide high temperature thermal energy (>900°C) is considered as a low carbon energy source to meet industrial heat demands [1]. The thermal energy storage (TES) systems are expected to increase the thermal energy system efficiency and to be bridge the gap between demand and supply of thermal energy [2]. Among the thermal energy storage methods, the chemical heat pumps (CHPs), which has advantages such as higher storage density and longer storage time without dissipation, is considered as an alternative to conventional sensible and latent TES system.

There are various kinds of thermochemical reaction working pairs for the CHP systems are under investigation according to the reaction temperatures, however, the appropriate thermochemical reactions for using at the high temperature thermal energy are limited. We introduced the lithium orthosilicate and carbon dioxide (Li<sub>4</sub>SiO<sub>4</sub>/CO<sub>2</sub>) reaction, Eq. (1), that has -127 kJ mol<sup>-1</sup> of reaction enthalpy at 700°C and proposed zeolite was as a CO<sub>2</sub> reservoir [3].

$$Li_4SiO_4(s)+CO_2(g) \square Li_2CO_3(s)+Li_2SiO_3(s), \quad (1)$$
  
$$\Delta H_r^{700^{\circ}C} = -127.0 \text{ kJ mol}^{-1}$$

Fig. 1 shows the schematic diagram of lithium orthosilicate/carbon dioxide/zeolite (Li<sub>4</sub>SiO<sub>4</sub>/CO<sub>2</sub>/Zeolite) CHP system. The decarbonation is an endothermic reaction corresponding to the thermal energy storage process, Fig. 1(a), and carbonation is exothermic reaction corresponding to the thermal energy output process, Fig. 1(b).



Fig. 1 Schematic diagram of Li<sub>4</sub>SiO<sub>4</sub>/CO<sub>2</sub>/Zeolite CHP system. (a) decarbonation/thermal energy storage process, (b) carbonation/thermal energy output process.

Although there were many studies for various CHP working pairs and theoretical operation for the whole temperature range, most of the investigations were carried out in powder state and there were few studies on large scale demonstrations. We developed a tablet form of Li<sub>4</sub>SiO<sub>4</sub> for practical using and designed two different reactors to demonstrate the thermal driving operation of a large scale high temperature CHP system without any mechanical power [4].

# **2.** Li<sub>4</sub>SiO<sub>4</sub> tablet and experimental apparatus *2.1 Materials*

A developed Li<sub>4</sub>SiO<sub>4</sub> tablet (K-tablet) was prepared for a lithium orthosilicate packed bed reactor (LPR). Lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>, 99.0%, Wako pure chemical industries, Ltd.), silicon dioxide (SiO<sub>2</sub>, 99.0%, Wako pure chemical industries, Ltd.), and potassium carbonate (K<sub>2</sub>CO<sub>3</sub>, 99.0%, Wako pure chemical industries, Ltd.) were prepared in 2.0:1.0:0.1 molar ratio as a precursor; K<sub>2</sub>CO<sub>3</sub> was added for enhancing the CO<sub>2</sub> absorption property. The precursor with chemical additives was compressed into the tablet ( $\Phi 6$  mm × 2 mm,  $\rho = 0.9$  g cm<sup>-3</sup>) under 10 MPa, then precast tablet was sintered at 785°C for 12 hrs under Ar atmosphere, Fig. 2.

The commercial zeolite (F-9, 1.40-2.36 mm, Wako pure chemical industries, Ltd.) was also introduced for zeolite packed bed reactor (ZPR).



Fig. 2 Image of produced K-tablet

#### 2.2 Reactors and demonstration system

The two reactors, LPR and ZPR (SUS 304), were designed in consideration of actual operating conditions. LPR works under high temperature, around 700°C, in a vacuum state and ZPR implements rapid temperature change in a specific time. Additionally, thermocouples were set in each reactor for measuring the temperature change during

the experiment.

The cross section of the vacuum chamber and the top view of LPR is shown in Fig. 3. The three alumina Tammann tubes (SSA-H-T8, OD × ID × L =  $50.0 \times 40.0 \times 180$  mm, Volume = 225 mL) with sheath heaters were used to configure the electric furnace. The electric furnaces were fixed in the vacuum chamber and the vacuum chamber was encased by ribbon and jacket heaters. In this study, 329 g of K-tablet was used for LPR. The ZPR consisted of two layers of the cooling tube and sheath heater on the inner and outer sides of the vacuum chamber for effective heating and cooling of zeolite bed, Fig. 4. The volume of the vacuum chamber is 4.9 liter and 3,300 g of commercial zeolite was filled into the reactor. Especially, metal mesh tube was positioned in the center of the reactor for enhancing CO<sub>2</sub> diffusivity.

The schematic diagram of the thermal driving operation system for  $\text{Li}_4\text{SiO}_4/\text{CO}_2/\text{Zeolite CHP}$  system is shown in Fig. 5. Two reactors were connected by flexible and rigid pipes with a solenoid valve, and mounted on an electric balance to measure the weight change during the experiment. The solenoid valve controlled supplying CO<sub>2</sub> pressure according to the set value. All temperature, pressure, and mass data

# were collected through the data acquisition system. *2.3 Experimental Procedure*

The thermal driving operation procedure could be divided into two processes for each reactor; carbonation of LPR/desorption of ZPR (CD experiment) corresponding to the thermal energy output process and decarbonation of LPR/adsorption of ZPR (DA experiment) equivalent to the thermal energy storage process.

Before initiating each experiment, the Li<sub>4</sub>SiO<sub>4</sub> tablet was fully decarbonated by heating at 715°C in a vacuum state, around 10 Pa. In the CD experiment, ZPR was heated up to 120°C for CO<sub>2</sub> desorption, then desorbed CO<sub>2</sub> was introduced to LPR with set pressure, 150 kPa, for 4 hours. After finishing the carbonation, close the main valve and cool down the ZPR for the DA experiment, around 2 hours. When the middle temperature of ZPR had been reached 15°C, the DA experiment carried out for 6 hours. In this study, 2 times of thermal driving operation were conducted; the one is single operation and the other is cyclic operation at same conditions.

(a)



Fig. 3 Image of (a) cross section of vacuum chamber and (b) top view of LPR

Fig. 4 Image of (a) cross section of vacuum chamber and (b) side view of ZPR

(b)



Fig. 5 Schematic diagram of thermal driving operation system

#### 3. Experimental results and discussion

Fig. 6 shows the thermal driving operation results of the developed Li<sub>4</sub>SiO<sub>4</sub>/CO<sub>2</sub>/Zeolite CHP system and each result showed similar temperature and mass change trends as follows.

- The middle temperature was sharply increased at the initial time of the CD experiment (thermal energy output process), on the other hand, the temperature was suddenly decreased at the beginning of the DA experiment (thermal energy storage process).

- The bottom temperature did not show particular fluctuation during the whole experiment. It is probably because of not enough  $CO_2$  diffusivity in the electric crucible.

- Upper temperature measured above K-tablet packed bed was lower than other temperatures and it represents an atmosphere of the electric crucible. The temperature was increased on the CD and DA both experiments by exothermic reaction and desorbed  $CO_2$  in high temperature respectively.

- The overall weight change of LPR and ZPR were corresponding well to each other and these results were used for estimation of thermal energy capacities.

The single operation (SO), Fig. 6(a), and first cycle of cyclic operation (1st CO) showed almost similar results. However, the highest temperature and mass conversion,  $\Delta m_{\text{carb}}$  [mg], in carbonation were decreased on the second cycle of cyclic operation (2nd CO) due to not enough decarbonation conversion,  $\Delta m_{\text{decb}}$  [mg].

The thermal energy output density,  $Q_{LPR}$  [kJ L<sup>-1</sup>], that is thermal energy released per unit volume of LPR was estimated at 331-395 kJ L-packed bed<sup>-1</sup> through the measured mass change.

#### 4. Conclusion

 $Li_4SiO_4/CO_2/Zeolite$  CHP system was investigated for utilizing the high temperature thermal energy generated from HTGRs. We developed the tablet form of  $Li_4SiO_4$  and designed two different types of reactors for demonstrating the thermal driving operation of the CHP system on a large scale.

Single and cyclic thermal driving operation was carried out and developed Li<sub>4</sub>SiO<sub>4</sub>/CO<sub>2</sub>/Zeolite CHP system showed a high possibility to utilize the high temperature thermal energy and enhance the thermal utilization efficiency of the high temperature gas cooled reactors.

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Fig. 6 Thermal driving operation results (a) single operation (SO), (b) cyclic operation (CO)

### A.3 Validation of Echo-PIV for inspecting inside of PCVs in 1F

Hiroshige KIKURA and Hideharu TAKAHASHI

#### 1. Introduction

The ongoing decommissioning of the Fukushima Daiichi (1F) nuclear power plant requires the inspection of the inside of containment vessels that have been submerged in water. These inspections must locate leaks and map the distribution of fuel debris in water with very low visibility. This study reports the design and testing of an echo-PIV system using DW-DAS imaging method that uses a single divergent signal wave (DW) and delay-and-sum processing (DAS) to efficiently map the interior and fluid flow within a submerged vessel.

#### 2. Design consideration

#### 2.1. Formulization of DW-DAS

A virtual point source is assumed behind the sensor array. Assuming that sound waves propagate from the virtual point source in concentric waves, the travel time at which the wave reaches each element is calculated from the distance and the speed of sound in the medium as the transmission delay,  $\Delta \tau$ :

$$\Delta \tau_e = \frac{\sqrt{(x_e - x_v)^2 + (y_e - y_v)^2}}{c}$$
(1)

Where x and y are the spatial coordinates, subscripts v and e distinguish the virtual source and the element respectively and c is the speed of sound in the medium.

The transmitted wave described above propagate in a diverging manner and cover a wide field of view with a single trans-mission. After receiving scatter echoes with all sensor elements at the same time, the echo intensity at each measurement point in the region of interest is calculated by summing the amplitudes of all received signals with the corresponding travelling time from the virtual source to the elements via the measurement point. This process describes the delay-and-sum algorithm. Finally, considering the decrease in amplitude over distance due to wave dispersion, the DW-DAS is formulated as follows:

$$I(x,y) = \left| \sum_{e=1}^{N} \frac{1}{\sqrt{d_v d_e}} H_e\left(\frac{d_v + d_e}{c}\right) \right|$$
(2)

Where *I* is the echo intensity, *N* is the number of elements, *H* is the Hilbert transform of the received echo signal, and *d* is the distance from the position (x,y).

#### 2.2. Echo-PIV processing

First, a DW transmission is repeated over the number of pulse repetitions at the pulse repetition frequency and all received signals are recorded. Then, the received signals are converted to echo images. The echo images are paired and those pairs are processed with a PIV algorithm to estimate the velocity vector field. Cross-correlation fast Fourier transforms (FFT) using a small window size with offsetting are used in the PIV algorithm. Estimated velocity vectors, are averaged over the number of image pairs and a final vector flow map is obtained. In addition, the averaged echo image is also recorded for measuring the shape of surface reflectors (e.g. debris).

#### 3. Feasibility tests

To validate the feasibility of echo-PIV using DW-DAS, an experiment mocking up a containment vessel containing simulated debris was conducted in the lab. In the experiment, a tank was filled with tap water and tracers were mixed in at certain density. A rough surfaced stone was placed at the bottom of the water tank to simulate debris. The water tank had an outflow outlet (Ø24 mm) as a leakage. Measurements were recorded while moving a used sensor array above the debris and leakage with a 5 mm pitch while draining the stored water from the outlet. The measurement conditions and imaging settings are shown in Table 1. The window sizes in the echo PIV algorithm were set as  $128 \times 128$ ,  $96 \times 96$ ,  $64 \times 64$  and  $32 \times 32$  pixels with a 50% overlap.

Table 1 Measurement condition.

Water	Sound	Flow	RIO	LIDE	LPF [MHz]	# of	# of	PRF
temp.	speed	rate	KIO []	INTI-1		image	meas.	
[°C]	[m/s]	[l/min]	[mm]	[MIIIZ]		[-]	[-]	[nz]
			$0 \le x \le 100$					
25	1497	3.0	$-40 \le y \le 40$	1.3	3.4	64	1	100
			Pitch 0.1					

#### 4. Results

Fig.1 shows the results of this mock-up test. The height corresponding to the maximum value of the averaged echo images at each measurement point was extracted and is displayed in grey-scale. The vectors displayed are only over 15 mm/s to facilitate visualizing the 3D flow behavior. As a result, the prototype system could identify the leakage point and determine the shape of the mock fuel debris. These results suggest that the DW-DAS echo-PIV method is applicable for sensing leakage points and underwater debris.

![](_page_23_Figure_23.jpeg)

![](_page_23_Figure_24.jpeg)

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### A.4 Extension of Measurable Velocity Limit in Ultrasonic Velocity Profiler by Wideband Phase Difference Method

Hiroshige KIKURA and Hideharu TAKAHASHI

#### 1. Introduction

Recently, an ultrasonic velocity profiler (UVP) is utilized on various flow, e.g. industrial piping flows, flows in food processing and two-phase (three-phase) flows. In our research group, the UVP method has been applied to Fukushima-daiichi nuclear power plant to detect a leakage point of contaminated water. the UVP method, however, has a measurable velocity limit; it is limited to utilize the UVP depending on flow conditions. Therefore, an extension of the measurable velocity limit in the UVP is contribute for a diffusion and a development of the UVP. We developed a velocity limit extension method, wideband phase difference method (WPD), as a novel flow velocity estimation method.

#### 2. Development of Measurement System

#### 2.1. Detectable Velocity Limit in UVP

An ultrasonic velocity profiler (UVP) can measure instantaneous velocity profiles by detecting Doppler frequencies on an ultrasonic beam path. To detect the Doppler frequency, the UVP method uses the pulse repetition method. The velocity information can be obtained from following equation.

$$v = \frac{J_{\text{PRF}} c}{4\pi f_c \sin \alpha} \cdot \Delta \theta , \qquad (1)$$

where *c* is the speed of sound in the medium,  $f_c$  is the ultrasonic basic frequency, and  $f_{PRF}$  is the pulse repetition frequency. The phase shift,  $\Delta \theta$ , is limited from  $-\pi$  to  $\pi$  by Nyquist theorem. Thus, the maximum measurable velocity along the flow direction,  $v_{limit}$ , is expressed as

$$v_{\text{limit}} = \frac{f_{\text{PRF}} c}{4f_{\text{c}} \sin \alpha} \,. \tag{2}$$

Therefore, a velocity aliasing is occurred when velocities over the  $v_{limit}$ .

#### 2.2. Wideband Phase Difference Method

To measure higher velocities, the wideband phase difference method (WPD) was developed as a de-aliasing method. This method employs the phase difference pattern within the signal bandwidth to modify the phase folding by the Nyquist theorem. Here, we consider the consecutive two echo signals,  $f_1(t)$  and  $f_2(t)$ , from the reflector. The second echo signal,  $f_2(t)$ , can be expressed as  $f_2(t) = f_1(t + \Delta t)$  due to the Doppler shift. Furthermore, the Fourier transforms of each echo signal are expressed as  $F_1(\omega)$  and  $F_2(\omega)$ , respectively.  $F_2(\omega)$  can be expressed as following equation using the relationship of  $f_2(t) = f_1(t + \Delta t)$ .

$$F_2(\omega) = e^{j\omega\Delta t} \cdot F_1(\omega). \tag{3}$$

Namely, the time shift of the echo signal in the time domain reflect to the phase shift in the frequency domain. The phase difference,  $\Delta \theta(\omega)$ , between the two echo signals is expressed as

$$\Delta \theta(\omega) = \arg[F_2(\omega) \cdot F_1^*(\omega)] = \omega \Delta t \qquad (4)$$

where the mark of \* means the complex conjugate. As Eq. (4), the phase differences in the frequency domain are proportional to  $\omega$  with  $\Delta t$  as a coefficient. The gradient coefficient,  $\Delta t$ , is preserved even if the aliasing is occurred. Therefore, we can get a higher velocity information from a phase difference pattern within the signal bandwidth.

#### 3. Demonstration of WPD Method

To verify the performance of WPD method, a piping flow measurement was carried out. Figure 1 shows the results of velocity profiler measurement in the horizontal pipe by WPD method. The velocity profiles over the conventional  $v_{limit}$  were obtained.

![](_page_24_Figure_21.jpeg)

#### 4. Conclusion

To develop the wide velocity range flow measurement system, UVP system implemented the de-aliasing method, WPD method, was constructed and the flow measurement in the horizontal pipe was carried out. As the result, the velocities over the conventional  $v_{limit}$  were obtained by WPD method.

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# A.5 Measuring bubble lengths in suppression pool experiment by ultrasonic pulse-echo measurement with tracking technique

Hideharu TAKAHASHI, Hideo NAGASAKA and Hiroshige KIKURA

#### 1. Introduction

During a nuclear reactor accident, the reactor vessel or containment failures are prevented by suppressing the pressure by condensation or by wetwell venting, which releases various gases into the environment after the radioactive material has been filtered out from it using a pool scrubbing effect. The pool scrubbing efficiency depends on the size of gas bubbles. However, there is only a limited knowledge on the bubble size and how it changes with different conditions.

To obtain experimental data, a suitable experimental technique is required. High-speed cameras (HSCs) are not applicable to large volumes of bubbly flow. Probes are intrusive and measure only at one point. In contrast, ultrasonic transducer (TDX) can easily go inside large fluid volume and conduct a not intrusive line measurement. However, available ultrasonic techniques are limited to single bubbles, which limits their application range. To extend the range a new technique capable of measuring multiple bubbles simultaneously was developed and its capabilities demonstrated.

#### 2. Methods

The pulse-echo (aka sonar) can be applied with two TDXs as shown on Fig. 1 to measure bubble lengths as d = x12 - x1 - x2. (1)

$$\begin{array}{c} x_{12} \\ x_1 \\ TDX1 \end{array}$$

Fig. 1 Basic configuration

0

The signal from each TDX was processed as in [1]. The processing consisted of two parts. The first part detected reflections from bubbles in the echo signal of each pulse repetition. The time it took the signal to reach the bubble and get back to the TDX shows the distance to the bubble surface ( $x_1$  for TDX1). The second part, tracking, connected the reflections from various pulse repetitions to form trajectories of bubble surfaces.

The technique was validated by comparing with a high-speed camera in a simplified experiment [2] and a low uncertainty of 0.3 mm was confirmed.

#### 3. Application to suppression pool environment

The environment of real suppression pools can reach up to 5 bar and a corresponding saturation temperature of water, which is higher than the operating range or regular TDXs. Resistant TDX were developed for this purpose and their performance was validated as shown in Fig. 2.

![](_page_25_Figure_15.jpeg)

#### 4. Bubble diameter data from scrubbing experiment

An experimental apparatus resembling a suppression pool was used. Mixture of steam and air were injected into a pool and the resistant TDXs were applied there to measure bubble length of each bubble. Bubble lengths were then converted to bubble diameters using a correlation form literature. The distributions of diameters were analysed and the effect of various parameters (water pool temperature, gas flow rate, injection diameter and radial position) was observed.

![](_page_25_Figure_18.jpeg)

#### 5. Conclusions

An ultrasonic pulse-echo technique suitable for bubble diameter measurement in a suppression pool experiment was developed. TDXs suitable for this condition were developed as well. Both were applied and the capability of the measurement system was demonstrated.

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# A.6 Fundamental Study of Sensing Technique Using MCF Rubber Sensor for Radiation Environment

Hideharu TAKAHASHI and Hiroshige KIKURA

#### 1. Introduction

Recently, MCF rubber mixed MCF (Magnetic Compound Fluid) with rubber such as silicone oil rubber or natural rubber was developed. MCF rubber is smart sensor by utilizing MCF compounding rubber, solidified by electrolytic polymerization. It is possible to self-sensing because it has self-induced voltage and it changes with normal force, shear force, etc. Furthermore, it is possible to develop as a wide variety of new elements such as capacitors or solar cells. In this study, we investigated the characteristics of MCF rubber under radiation environment.

#### 2. Making of MCF rubber sensor

The solidification model for MCF rubber using electrolytic polymerization method is shown in Fig.1. Anionic polymerization occurs at the cathode and cationic polymerization at the anode. Our used MCF rubber liquid consisted of 3 g carbonyl Ni powder, with particles of the order of  $\mu$ m having bumps on the surface (No. 123, Yamaishi Co., Ltd., Noda, Japan), 0.75 g water-based MF with 40 wt% Fe<sub>3</sub>O<sub>4</sub> (W-40, Ichinen-Chemicals Co., Ltd., Shibaura, Japan), 1.5 g dopant material, 3 g of NR-latex (Rejitex Co., Ltd., Atsugi, Japan).

At first, the liquid was poured between stainless-steel plates with a 1 mm gap, a constant electric field was applied at 6 V, an electric current of 2.7 A was passed between the plates for 10 min, and a 312 mT magnetic field was applied by permanent magnets as paired opposites across the liquid.

![](_page_26_Figure_9.jpeg)

Fig. 1 Model of electrolytic polymerization and solidification of the MCF rubber.

#### 3. Result and discussions

We studied the relationship between the potential and current under  $Co_{60}$   $\gamma$ -ray irradiation. Fig.2 shows the time course of the rest potential under irradiation, and Fig.3 shows CV (Cyclic Voltammetry) diagram of MCF rubber under  $Co_{60}$   $\gamma$ -ray irradiation. The label "on" and "off" represent the time at which the irradiation started and stopped, respectively. A fast increase and decrease in the potential shown as "on" and "off".

It was suggested that it could be applied as a battery or detector for radiation.

![](_page_26_Figure_14.jpeg)

Fig.2 Time course of electric potential of the MCF rubber under radiation environment.

![](_page_26_Figure_16.jpeg)

#### Reference

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### **A.**7

### Material Compatibility Study

### on Liquid Metal Divertor of Fusion Reactors

Masatoshi KONDO

#### 1. Introduction

Fusion reactor is an innovative power plant, which is necessary for a sustainable society. The divertor is an important component, which is responsible for power exhaust and impurity removal through guided plasma exhaust in magnetic confinement fusion reactors. The divertor is exposed to high-energy neutrons and large heat load about 5 to 10 MW/m<sup>2</sup>. The critical issue for the development of the divertor is to mitigate the damage of its structural material by the large heat load.

The design study on solid divertor is being conducted for the development of fusion DEMO reactors. The liquid surface divertor, in which the surface of the structural material is covered and protected by the liquid metal coolant, is also being proposed. In this concept, the recycling of plasma particles may be suppressed due to their adsorption and transport by the flowing liquid metal coolant. Therefore, the plasma performance is also improved. Thus, the liquid divertor concepts have some strong advantages, which can achieve longer lifetime of the divertor material and better plasma performance. Liquid tin (Sn) is a promising coolant of the liquid surface divertor. Figure 1 shows a schematic diagram of the liquid Sn surface divertor [1]. US-Japan collaboration on the liquid divertor study was started as TASK 3(Reaction dynamics between solid material-liquid metal interfaces for liquid divertor systems) of FRONTIER project [2] in FY2019. The important issue is chemical compatibility between liquid Sn and structural materials. Thus, corrosion characteristics of candidate structural and functional materials in liquid Sn were investigated in the present study.

# 2. Thermophysical and chemical properties of liquid metal tin (Sn)

#### 2.1. Thermophysical properties of liquid Sn

Liquid metal Sn has thermophysical properties as summarized in Table 1. Its atomic number is 50. The density of liquid Sn is large, though it is smaller than that of liquid Pb. The melting point of Sn (i.e. 505.1) is higher than alkali metals (e.g. Na: 370.9K and Li: 453.7) and lower than heavy metals (e.g. Pb: 600.7K and Bi: 544.7K). The boiling point of liquid Sn is very high. The thermal conductivity of liquid Sn is 33 W/(mK) and its Pr number is very small. As such, liquid Sn is excellent coolant for energy plants operated at high temperature. The vapor pressure of liquid Sn is extremely low rather than the other liquid metals [3]. This is a strong motivation to adopt liquid Sn as liquid divertor coolant, since its small evaporation does not affect on a plasma performance.

#### 2.1. Checmial properties of liquid Sn

The oxygen potential of liquid Sn and Gibbs free energy for formation of various oxides at 800K is summarized in Fig.2. The oxygen potential of liquid Sn is estimated in equation;

$$\phi_{0,\text{Sn}} = \frac{1}{2} \Delta G_{f,\text{SnO}_2} + RT \ln(\frac{c}{cs}) \quad [\text{kJ/mol}] \qquad (1),$$

where  $\Delta G_{f,SnO2}$  is Gibbs free energy for formation of SnO<sub>2</sub>, R is gas constant, T is temperature, C is oxygen concentration, and Cs is oxygen solubility of liquid Sn. C/Cs is assumed to be 0.01 in Fig. 2. The oxygen potential of liquid Sn is rather high. Though H<sub>2</sub>O reacts with Sn to form SnO<sub>2</sub>, various oxides can survive in liquid Sn from the view point of thermodynamic stability.

![](_page_27_Figure_17.jpeg)

Liquid Sn coolant loop

Fig.1 Schematic diagram of liquid Sn surface divertor [1]

#### 2. Corrosion test with various test materials

#### 2.1. Experimental conditions

Table 2 presents the list of the materials used for the current corrosion tests. JLF-1 is a reduced activation ferritic martensitic steel, and this is one of the promising structural materials for the liquid divertor. The corrosion resistance of Al-rich steels which can form Al-rich protective oxide layer were also investigated. They were oxidized in air at 773K or 1073K to form the oxide layer on their surface [4] before the

immersion to liquid Sn. Pure W has a melting point of 3695K. The combination of liquid Sn coolant and W wall may promote the reliability of the liquid divertor concept. The corrosion characteristics of pure W and its sintered compacts in liquid Sn were also investigated. SiC and other metals such as Zr, Ti, and Y which have revealed their corrosion resistance in various liquid metals were also tested in the present study.

Corrosion tests were performed at 773K for 262 and 250 hours. A rectangular specimen was placed in a static Sn. The crucible was made of 316L austenitic steel. The inventory of liquid Sn was approximately 3cc. After the corrosion tests, specimens were picked up from liquid Sn. The surface and the surface cross section of the specimens were metallurgically analyzed by FE-SEM/EDX, EPMA and AES without a removal of adhered Sn on the specimen surfaces.

![](_page_28_Figure_3.jpeg)

Fig. 2 Thermodynamic stability of liquid Sn at 800K (C/Cs=0.01)

#### 2.2. Experimental results

Intermetallic compounds of Fe and Sn were detected at the interface between JLF-1 and liquid Sn [1, 3]. The thickness was approximately 20 $\mu$ m and it was thinner than that formed in liquid Sn at 873K [4]. These results indicate that the temperature dependency of this alloying corrosion is large. Similar corrosion was observed on the surface of JLF-1 specimen which was pre-oxidized and covered by Fe-rich oxide layer. The Fe-rich oxide layer could not function as a corrosion barrier, since the layer reacted with Sn and formed unstable oxides.

The Al-rich steels revealed corrosion resistance due to the formation of Al-rich or Cr-rich layers on their surface. These layers did not react with liquid Sn. The alloying reaction between steels and liquid Sn were totally suppressed.

The specimen of Pure W bulk and W sintered compacts revealed corrosion resistance, since the solubility of W in liquid Sn was quite low. The diffusion of Sn along the boundaries of the sintered matrix was detected in the W sintered compact, since Fe, Cu and Ni were used as a sintering dopant and reacted with liquid Sn. The surface of the W bulk and the sintered compacts were carbonized in liquid Sn. Liquid Sn must contain certain amount of carbon though Sn does not form its carbide. The carbon dissolved in liquid Sn reacted with W and formed the carbide layer on the specimen surface. AOD-HAF1 which was protected by carbonization layer revealed excellent corrosion resistance.

#### 3. Conclusions

Major conclusions are follows;

- 1. The reduced activation ferritic steel JLF-1 corroded in liquid Sn through the formation Fe-Sn intermetallic compounds.
- 2. The Al-rich steels revealed corrosion resistance in liquid Sn, since the alloying reaction was suppressed by the Alrich or Cr-rich oxide layers.
- 3. Pure W and the sintered compacts revealed corrosion resistance, since the solubility of W in liquid Sn was quite low.

#### Acknowledgment

The author acknowledges Dr. N. Ohno for her support on the corrosion study on FeCrAl-ODS steel SP10. The author acknowledges Dr. B. Pint, Dr. J. Jiheon at Oak Ridge National Laboratory and all the members of US-Japan collaboration research FRONTIER project TASK-3 for their fruitful discussions. The author acknowledges Mr. T. Ohe, Mr. J. Nakano and Mr. T. Awaji at Marumoto Struers K. K for their technical advice on the cross sectional polishing procedure of the JLF-1 specimen and the solidified Sn.

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Table 1 Thermophysical properties of liquid Sn and other liquid metals

	Melting point (K)	Boiling point (K)	Density at 800K (kg/m <sup>3</sup> )	Pr
Sn	505.1	2543	6761	0.0094
Li	453.7	1615	481	0.027
Pb	600.7	2010	10360	0.018

		Cr	Al	W	Ti	Ni	Si	С	Mn	Others	Fe
316L		17.3	-	-	-	12.3	0.54	0.018	1.6	2.2Mo, 0.028P	Bal.
JLF-1	RAFM	9	-	1.94	-	-	-	0.09	0.49		Bal.
Fe <sub>2</sub> O <sub>3</sub>	- Sintered compact	-	-	-	-	-	-	-	-	300	70
Cr <sub>2</sub> O <sub>3</sub>		68.4	-	-	-	-	-	-	-	31.60	-
NTK04L [2]		16.33	1.04	-	-	7.16	0.25	0.08	-	0.029P	Bal.
FeCrAl- ODS(SP10)	Al-rich steel	17.84	3.34	-	0.3	-	0.41	-	0.14	0.41Si	Bal.
NAS		14.76	6.40	-	0.5	-	-	-	-	0.47Y <sub>2</sub> O <sub>3</sub> , 0.37Zr, 0.22Ex.O	Bal.
Al <sub>2</sub> O <sub>3</sub> bulk	Sintered compact	-	53	-	-	-	-	-	-	470	
HAC2	- Sintered compact	-	-	94	-	4	-	-	-	2Cu	-
HAF2		-	-	93	-	5	-	-	-	-	2
HAF-1 AOD	Carbonization	-	-	97	-	2	-	-	-	-	1
Pure W	-	-	-	100	-	-	-	-	-	-	
SiC		-	-	-	-	-	70	30	-	-	-
ZrO <sub>2</sub> /Zr	Surface oxidized	-	-	-	-	-	-	-	-	-	-
Zr		-	-	-	-	-	-	-	-	100Zr	-
Ti	Pure metal	-	-	-	100	-	-	-	-	-	-
Y		-	-	-	-	-	-	-	-	100Y	-

Table 2 Thermophysical properties of liquid Sn and other liquid metals (wt%)

**B.** Actinide Management Division

## B.1 Separation activities of Takeshita-lab in FY2018

Masahiko NAKASE, Wu HAO, Sayumi ITOH, Nao TSUTSUI, Tatsuro MATSUMURA and Kenji TAKESHITA

#### 1. Introduction

In Takeshita Lab, we are developing ligands and sorbents for separation of various metal ions such as Miner Actinide (MA), Platinum Group Metal (PGM) and Cs, Sr which are important in the field of nuclear fuel cycle. In FY of 2018, we have developed some of the ligands and characterization was done by several analytical method such as solvent extraction, UV-vis titration, single crystal XRD and Extended X-ray Absorption Fine Structure (EXAFS) experiments. The annual achievement related to ligand development in FY2018 was briefly reported.

#### 2. Development of Ligands for separation of metal ions

Separation of f-block element is especially important for advanced nuclear fuel cycle and its reprocessing. To separate MA from Ln, we reported some of the N,O-hybrid donor ligands as shown in Figure 1 [1]. The alkyl-substituted 1,10-phenanthroline-2,9-diamide (PTDA) and 1,10-phenanthroline-2-carboxamide (PTA) were either synthesized or purchased from a company. The extraction results suggested that the solvent affected greatly on the extraction property. The synchrotron experiments were effective to investigate the structure of complexes in the actual solvents (nitrobenzene and n-dodecane). The EXAFS experiments were conducted at Super Photon Ring-8 GeV (SPring-8) for K1 edge of Ln and Aichi synchrotron center, SAGA Light Source and Photon Factory for L<sub>3</sub> edge of Ln. The theoretical fittings to investigate the coordination number, atomic distance, Debye-Waller factor and energy shift ( $\Delta E_0$ ) were calculated based on the crystal structure obtained in ethanol solution (for example as shown in Figure 1(2)). In addition, extraction experiments with Am and Cm were done with the PTDA derivatives at Nuclear fuel Cycle safety Engineering research Facility (NUCEF) of Japan Atomic Energy Agency (JAEA) and contactor work was also continued [2]. The novel ionic liquid compounds were also synthesized and the extraction properties of Ln was reported as shown in Figure 2. The ligand-immobilized thermosensitive hydrogels for f-element recognition was characterized by EXAFS[3, 4].

![](_page_32_Figure_8.jpeg)

Figure 1 Structure of (1)PTDA and PTA and (2)crystal

![](_page_32_Figure_10.jpeg)

Figure 2 Structure of TPEN-type ionic liquid

Not only extraction of f-lement, but also PGMs extraction from nitric acid was performed. Figure 3 is the example of the extractant for PGMs separation developed in Takeshita Lab by the collaboration with National Institute of Advanced Industrial Science and Technology (AIST). The new form of complex (Pd : Ligand = 1 : 2) was confirmed by loading experiments and IR spectra [5].

![](_page_32_Figure_13.jpeg)

Figure 3 *N*,*N*'-dimetyl-*N*,*N*'-ditholyldiglucolamide (MTTDGA)

#### 3. Conclusion and plan

In FY2018, we reported insights about separation of various metal ions. The further results and information in detail can be obtained in the literatures in reference. We will expand our activities of mechanism study on ion recognition and collaboration with JAEA will be continued.

#### Acknowledgment

The activities in this report were supported by some JSPS grant-in-aid and collaborative work with JAEA (see the list of collaborative researches in the bulletin, FY2018)

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# **B.2** Study on nuclear fuel cycle with less-impact radioactive waste management -Impact of Cs and Sr separation on reducing the geological disposal area

Tomohiro OKAMURA, Eriko MINARI, Masahiko NAKASE, Hidekazu ASANO

and Kenji TAKESHITA

#### 1. Introduction

The management of high-level radioactive waste (HLW) is indispensable and one of the main issues for the steady use of nuclear energy. In recent years, R&D of partitioning and transmutation (P&T) technologies have been promoted for the purpose of reducing the amount of HLW, geological disposal area and potential radiotoxicity. The nuclear fuel cycle (NFC) is composed of multiple sectors such as reactor operation, reprocessing of spent nuclear fuels (SNFs), vitrification and geological disposal. Therefore, it is important to maximize the merits of P&T for reducing the burden of HLW management with considering all the sectors. For the first step, we focused on Cs and Sr that strongly contribute to heat generation of vitrified waste, and examined how the separation of Cs and Sr affects the geological disposal area under the various conditions of NFC.

#### 2. Methodology and condition

Geological disposal area per electricity generation  $(m^2/TWh)$  was used as an index for quantitative evaluation of the NFC to comprehensively consider both front-end and back-end from viewpoint of the geological disposal [1]. This index can evaluate the a minor fluctuation of number of vitrified waste which can be changed by conditions of NFC. In addition, CAERA (Comprehensive Analysis of Effects on Reduction of Disposal Area) index (kg/m<sup>2</sup>) currently reported by our group was also utilized [2]. In this evaluation, it separation of Cs and Sr from HLW generated from spent UO<sub>2</sub> fuels was evaluated with varying the condition as followings; 1 the initial fuel composition, 2 fuel burnup, 3 cooling period of SNFs, 4 waste loading of vitrified waste, 5 storage period of vitrified waste and 6 emplacement methods of waste package.

#### 3. Result and Discussion

The following 4 conditions of the NFC related to the effect of Cs and Sr separations on the reduction of geological disposal area were briefly summarized as followings;

1. Fuel burnup:

The elemental composition in the vitrified waste was changed depending on the fuel burnup. The higher fuel burn-up condition enhanced effect of Cs and Sr separation. 2. Cooling period of SNF

The elemental composition in the vitrified waste was greatly changed by cooling period of SNFs. In order to obtain the maximum effect of Cs and Sr separations, the

cooling period of SNF should be as short as possible.

3. Waste loading in vitrified waste

The extent of waste loading into vitrified waste was related to the number of vitrified waste. The higher waste loading made more reduction of the number of vitrified waste, and the heat generation of vitrified waste was correspondingly increased. In such a case, Cs and Sr separations were effective with extended cooling period of SNFs before the reprocessing.

4. Storage period of vitrified waste

The storage period of the vitrified waste is the period from vitrification to geological disposal. The storage period of vitrified waste and separation of Cs ad Sr have huge impact on reduction of the heat generation of vitrified waste in relatively early period of geological disposal. In case with separation of Cs and Sr, our calculation revealed that approximately 83% of geological disposal area can be reduced. The further investigation and results were summarized the reference materials [1,2].

#### 4. Future Work

In FY2018, we expanded the scenario study on NFC to understand how the nuclide separations affect the area for final geological disposal. Not only Cs and Sr separations, but also the minor actinide separations were studied in this year. The results were reported at domestic and international conferences as well as publication (6 presentations at domestic conferences, 1 presentations at international conferences and 1 paper have been published). Also, we newly started the development of NFC simulation code under the supervision of Dr. Nishihara in Japan Atomic Energy Agency. The Nuclear Material Balance (NMB) code to further investigate the optimum NFC scenario. In addition, one of the student (Ms. Minari) in Takeshita-lab studied for three months at International Atomic Energy Agency. She engaged in the upgrade of the Nuclear Fuel Cycle Simulation System (NFCSS). Her work was highly evaluated at the session and the further collaboration will be continued. The optimization and scenario studies of NFC were conducted by externally funded by Radioactive Waste Management Funding and Research Center (RWMC) and the collaboration will be expanded in next year.

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#### Acknowledgment

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### **B.3** Thermoresponsive Polymer Phase Transition-Based Direct Extraction of Platinum Group Metals from Nitric Acid Solutions

Haruka TATENO, K.C. PARK, and Takehiko TSUKAHARA

The separation of platinum group metals (PGMs) such as ruthenium (Ru), rhodium (Rh), and palladium (Pd) from high-level liquid waste (HLLW) is critical essential, because these metals cause problems for vitrification processes, e.g., instability of the glass melter and defects of the vitrified glass. Recently, we have proposed an environmentally-friendly and riskless separation technique of metal ions using thermosensitive polymer as an alternative to a conventional solvent extraction which consumes large amounts of toxic and inflammable organic solvent [1-3]. Poly(N-isopropylacrylamide) (PNIPAAm) is a typical thermosensitive polymer which can change the hydrophilicity and hydrophobicity across the lower critical solution temperature (LCST) at around 32 °C in water. When PNIPAAm and a hydrophobic ligand in an aqueous solution containing target metal ions are mixed below LCST, the metal-ligand complex can be formed in the water-soluble PNIPAAm. By increasing temperature above LCST, PNIPAAm undergoes gelification and the metalligand complex is simultaneously absorbed onto the PNIPAAm gel by hydrophobic interaction. As a result, the target metal can be directly extracted in the PNIPAAm aggregates from the aqueous solution (Fig. 1). In the present study, we applied this technique to extract PGMs from nitric acid solutions like HLLW.

The extraction experiments of  $Ru^{3+}$ ,  $Rh^{3+}$  and  $Pd^{2+}$  were carried out in nitric acid solutions ([HNO<sub>3</sub>] = 0.0–2.0 M). The solution of target metal ion(s) containing PNIPAAm and ligand, 1,3-dibutylthiourea (DBU), was vigorously stirred at the temperature below LCST for 1 hour. Subsequently, gelification of PNIPAAm was performed by vigorous stirring above LCST for 1 hour. After the gel adhered onto a stir bar and the wall of beaker, the remaining aqueous solution was recovered and diluted. The concentrations of  $^{101}Ru$ ,  $^{103}Rh$  and  $^{106}Pd$  contained in the diluted solutions were measured by inductively coupled plasma-mass spectrometry.

The effects of DBU concentrations on extraction percentages (E%s) of Ru<sup>3+</sup>, Rh<sup>3+</sup>, and Pd<sup>2+</sup> were examined in 0.0–2.0 M HNO<sub>3</sub> solutions. The results are shown in Fig. 2. We found that the extractability increased with increasing [DBU], and reached each maximum value by adding more than 5 ~ 10 folds of DBU relative to the amount of each metal ion. Pd<sup>2+</sup> was attained 100% by the addition of DBU regardless of [HNO<sub>3</sub>]. The E% was reached to 40 % and 56 % at [HNO<sub>3</sub>] = 0.1 M and [HNO<sub>3</sub>] = 0.0 M for Ru<sup>3+</sup> and Rh<sup>3+</sup>, respectively, and then decreases with increasing [HNO<sub>3</sub>]. Therefore, we determined the extraction stoichiometries with slope analysis method. The extracted species of Ru<sup>3+</sup> and Pd<sup>2+</sup> were found to be 1 : 1 (metal : DBU), and 1 : 1 or 1 : 3 complex depending on the [DBU] concentrations, respectively. On the other hand, the

slope of 0.3 was obtained for  $Rh^{3+}$ . This fact suggests that not only  $Rh^{3+}$ –DBU complex but also other species such as  $Rh^{3+}$ -aquo complex adsorbed to the amide moiety of PNIPAAm are co-extracted, because  $Rh^{3+}$  has fairly high hydration energy. Moreover, the concentration dependence of NaNO<sub>3</sub> against *E*%s of  $Ru^{3+}$  and  $Rh^{3+}$  was examined in HNO<sub>3</sub> solution with the constant  $[H^+] = 0.1$  M. The results showed that the *E*%s of each ion were increased with increasing  $[NO_3^-]$ . Such tendencies indicate that  $H^+$  and  $NO_3^-$  of HNO<sub>3</sub> yield opposite effects on the *E*%s, *i.e.*,  $H^+$ reduces the adsorption capacity due to the protonation of amide groups of PNIPAAm, but NO<sup>3-</sup> facilitates the formation of metal–DBU complex due to the hydration breaking.

![](_page_34_Figure_8.jpeg)

Figure 1. Conceptual illustration of the phase transition-based gelification method.

![](_page_34_Figure_10.jpeg)

Figure 2. The effect of ligand concentration on the extractability in each HNO<sub>3</sub> solution. Conditions; extraction temperature 60 °C, extraction time 1 hour.

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### **B.4** Visual Sensing of Metal Ions by On-Chip Photonic Crystal Polymer

Kaname SAGA, Chen SHEN, K.C. PARK, and Takehiko TSUKAHARA

For decommissioning of the Fukushima Daiichi Nuclear Power Station, the chemical analysis of radioactive wastes has become one of the important issues. Since the wastes include wide variety of chemical compositions and various radionuclides, general analysis methods which require complicated and time-consuming chemical operations are often inadequate. Accordingly, novel rapid and simple analysis method of metal species is expected to be established. Our previous studies have found that poly-N-isopropylacrylamide thermoresponsive (PNIPAAm) copolymers with functional ligands make it possible to adsorb and desorb metal ions selectively by just temperature swing across the lower critical solution temperature [1,2]. Therefore, in the present study, we aimed to develop a portable colorimetric micro sensing device, in which target metal ions could be detected by the diffraction wavelength shifts of PNIPAAm-gel enclosing photonic crystal (PC) according to Bragg-Snell law [3].

Core-shell nanoparticles consisting of PNIPAAm, polystyrene, and N-donating 1-vinylimidazole (VI) ligand were synthesized by two-step free radical and emulsion polymerization methods. A micro dam structure was fabricated in a thin PDMS film coated on a glass substrate using SU-8 photoresist mold, and a disk-shaped polyacrylamide hydrogel was placed onto the PDMS dam structure. And then, a suspension containing the core-shell nanoparticles was dropped and dried on the hydrogel, resulting in the formation of poly(NIPAAm)-VI copolymer-based PC due to self-assembly of the core-shell nanoparticles. After the PC was enclosed by PDMS plate and the PDMS chip enclosing PC was peeled from the glass substrate, the PDMS chip was bonded with a PDMS-microchannel. The fabrication procedures and fabricated chip are shown in Figure 1.

![](_page_35_Figure_6.jpeg)

Figure 1. Fabrication procedures and fabricated chip.

Molecular structures and sizes of the synthesized core-shell nanoparticles were characterized by using attenuated total reflection infrared spectroscopy and dynamic light scattering, respectively. The results showed that the 100 nm-sized particles drastically decreased with increasing temperature and by adding cations (lanthanides) and anions. Therefore, in-situ sensing cation species was demonstrated according to changes of color and diffraction spectra of poly(NIPAAm)-VI copolymer-based PC. Although the dried poly(NIPAAm)-VI copolymer-based PC had typically dark-blue color, by introducing pure water into the microchannel, the nanoparticles were swollen and the PC color was shifted to greem-yellow. In this case, the diffraction spectra also showed that the apparent peak shift from the wavelength of about 460 nm to 560 nm. When aqueous solutions containing trivalent lanthanide ions (lanthanum ( $La^{3+}$ ) or lutetium ( $Lu^{3+}$ )) were introduced into the microchannel, the colors were changed to bluish-green and metallic-blue for La<sup>3+</sup> and Lu<sup>3+</sup>, respectively. Such color changes corresponded to 25 nm and 40 nm blue shifts. These results indicate that the micro sensing device can be potentially used for rapid screening analysis of metal ions in radioactive waste solutions.

![](_page_35_Figure_9.jpeg)

Figure 2. Time-dependence of color and reflection spectrum changes of poly(NIPAAm)-VI copolymer-based after the addition of lanthanide ions.

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C. Global Nuclear Security Division

## **C.1**

# An innovative fast reactor core design for rapid reduction of separated Pu and its proliferation concerns

#### Rie FUJIOKA, Hiroshi SAGARA and Chi Young HAN

#### 1. Overview

An innovative FR core design was proposed to enable the rapid reduction of separated Pu physically by nuclear transmutation and managerially by conversion to an irradiated form, thereby downgrading its category in the nuclear security and safeguards regulations. The design also facilitated the equivalent or, in some cases, even better operation and safety performance compared to a typical FR, as determined by numerical neutronic analysis. The impact of the innovative core design for the rapid reduction of separated Pu was evaluated on mass balance and non-proliferation features, and compared to separated Pu storage on the ground or rapid reduction of separated Pu in the FR or the LWR.

Firstly, to find the fundamental characteristics of the FRs for rapid reduction of separated Pu, four core designs of FRs based on the demonstration type fast reactor, JSFR, were evaluated and compared in terms of Pu loading and Pu reduction. It was revealed that a core design with the blanket fuel regions removed and a higher-enriched loading of MOX fuel in core regions caused the highest Pu loading and Pu reduction ratio. These are preferable characteristics for rapid reduction of separated Pu, however the key issues of such core design were initial excess reactivity, positive void reactivity, and peaking factor.

Secondly, based on the results of fundamental characteristics analysis, the FR core design and specification for rapid reduction of separated Pu was derived for multi-parametrical surveillance, and provided the equivalent or better operation and safety performance than the typical FR. In order to overcome the key issues, three approaches were comprehensively examined; the age impact of MOX fuel was very significant to the initial excess reactivity compensation to accept a high initial 17.5 t Pu loading and 0.17 Pu reduction ratio. Placement of the gas plenum in the axial blanket

led to the improvement of the void reactivity, and the Pu enrichment zoning gradually became denser in the outer core which led to an improvement of the peaking factor, comparable to the requirements of a typical FR.

Finally, mass balance and non-proliferation features were evaluated and compared between separated Pu storage as the MOX powder form in containers, separated Pu reduction by MOX fuel irradiation in that proposal core of the FR and separated Pu reduction by MOX fuel irradiation in the typical PWR type of LWR, with 47.9 t separated Pu equivalent in Japan. In terms of mass balance, the amount of separated Pu storage on the ground would never change via separated Pu reduction in LWR, or FR would be completed within a finite period of time. Compared with MOX fuel irradiation, all Pu reduction time in one unit of FR was 19.5 years, which implies that the reduced speed in FR was 4.4 times faster than that in LWR due to the higher initial Pu loading, and total accumulated weight of Actinides and FP in the irradiated MOX fuel assemblies in FR was 40% smaller than those in the LWR. In terms of non-proliferation features, the nuclear material categories in the nuclear and safeguards regulations security were evaluated; MOX powder form in containers was the highest grade and the strictest regulations would be maintained in both the nuclear security and safeguards. On the other hand, the Categories of MOX fuel irradiated in the FR or the LWR would be downgraded in 1-step and the same as those of conventional spent nuclear fuel that exists abundantly worldwide.



Fig. 1 Categorization of non-proliferation features in Pu management options

#### Reference

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## C.2 Effect of Al-B-C Additives on Microstructure of Porous SiC Ceramics and Their Properties

Katsumi YOSHIDA, Anna GUBAREVICH and Toyohiko YANO

#### 1. Introduction

Silicon carbide (SiC) ceramics have been recognized as an attractive material for structural applications such as aerospace industries, nuclear and fusion applications and high-temperature gas turbine because it shows excellent properties such as high thermal and chemical stability, high stiffness, high hardness, high thermal conductivity, excellent oxidation, corrosion and wear resistance, high mechanical strength at high-temperatures, low thermal expansion, and good resistance to high-energy neutron irradiation. Recently, porous ceramics have received great attention in many fields. Porous ceramics have been traditionally used as heat insulating materials and refractories. Furthermore, porous ceramics have been applied as catalyst carrier, diesel particulate filter (DPF), membranes for gas separation, filters for water purification, adsorbent, electrodes for batteries, and bone-graft substitutes and so on. In order to extend the application of porous ceramics, their pore size control and surface functionalization have been required. Present authors have proposed the surface functionalization of porous ceramics based on in-situ grain growth [1,2]. Figure 1 shows our concept of porous ceramics based on in-situ grain growth. The interesting structures seen in nature such as calcareous caves have reached our concept regarding the surface functionalization of porous ceramics. In-situ grain growth for porous ceramics proposed in our studies has been defined as the grain growth and the grain orientation on the inside of pore walls and the frameworks of porous ceramics during sintering, and we have tried to fabricate porous ceramics based on in-situ grain growth.

In this study, porous SiC ceramics with in-situ plate-like grain growth were fabricated by pressureless-sintering using aluminum (Al), boron (B) and carbon (C) additives and the effects of these additives on the microstructure of porous SiC ceramics and their properties were investigated.

#### 2. Experimental procedures

#### 2.1. Fabrication of porous SiC ceramics

Submicron-sized  $\beta$ -SiC powder (average particle size; 0.32 µm) was used as the starting material, and aluminum (Al), boron (B) and carbon (C) were added to SiC powder as the sintering additives. In this study, aluminum nitrate (Al(NO<sub>3</sub>)<sub>3</sub>•9H<sub>2</sub>O) was used as the Al source. The composition of B and C was fixed as 0.15 and 1.0 wt%, respectively. The composition of Al was changed from 0.03 to 2.0 wt%. The amount of Al-nitrate addition was determined to be the composition of metallic aluminum described above. Beta-SiC powder and sintering additives were mixed by wet ball-milling for 24 h using SiC balls, and then dried with a rotary evaporator. The dried powder



Fig.1 Our concept of porous ceramics prepared with in-situ grain growth

was crushed with SiC mortar and pestle, followed by sieving with mesh with opening of 300 µm. Polymethyl methacrylate (PMMA) spheres with particle size of 30 µm was used as the pore former. Spherical PMMA particles were added to the mixture of SiC powder and sintering additives. The amount of spherical PMMA particles was 17 wt% (volume fraction of PMMA to SiC powder was 40 vol%). The powder was mixed using SiC mortar and pestle. After mixing, the powder was sieved through mesh with opening of 300 µm again. The sieved powder was formed into cylindrical compacts under a uniaxial pressure of 17 MPa followed by heat-treatment at 400°C for 1 h in air at a heating rate of 1 °C/min in order to remove the PMMA particles in the compacts. The compacts were put in a graphite crucible with a lid, and then the compacts were pressureless-sintered at 2150°C for 30 min in argon flow. From room temperature to 1500°C, the compacts were heated at a rate of 30 °C/min under vacuum and held for 30 min. Subsequently argon gas was introduced into the furnace, and then the compacts were heated at a rate of 30 °C/min to 2150°C. For comparison, porous SiC ceramics with only B and C additives were fabricated by the same process described above.

## 2.2. Characterization, mechanical and thermal properties of porous SiC ceramics

Bulk density ( $\rho$ ) and open porosity of porous SiC ceramics were measured by the Archimedes' method, and their pore size distribution was measured by mercury intrusion method using the mercury porosimeter. Crystalline phases of the porous SiC ceramics were identified by a powder X-ray diffraction analysis and the fractions of SiC polytypes in the porous SiC ceramics was

determined by the Ruska's method. Their microstructure was observed with a field-emission-type scanning electron microscope (FE-SEM). Compressive strength test was conducted at room temperature using cylindrical porous SiC ceramics (12 mm in height and 5 mm in diameter). Thermal diffusivity ( $\alpha$ ) and specific heat ( $C_p$ ) were measured by laser flash method at room temperature. Thermal conductivity of the porous SiC ceramics was calculated by multiplying  $\rho$ ,  $\alpha$  and  $C_p$ .

#### 3. Results and discussion

#### 3.1. Bulk density and open porosity

Bulk density and open porosity of porous SiC ceramics sintered with B-C additives were 1.67 g/cm<sup>3</sup> and 47 %, respectively. Bulk density of porous SiC ceramics with Al-B-C additives ranged from 1.44 to 1.92 g/cm3, and their open porosity was varied from 37 to 53 %. At 0.1 wt% Al additive or less, the porous SiC ceramics sintered with Al-B-C additives showed lower bulk density and higher open porosity than the porous SiC ceramics with B-C additives. Bulk density of the porous SiC ceramics sintered with more than 0.1 wt% Al additive increased to 1.8 -1.9 g/cm<sup>3</sup>, and their open porosity decreased to 37 - 39 %. Furthermore, bulk density of the porous SiC ceramics increased and their open porosity decreased up to 0.5 wt% Al additives due to the improvement of their sinterability. However, Al additive above 0.5 wt% did not increase the bulk density of the porous SiC ceramics. These results suggested that 0.5 wt% Al additive or more with B-C system enhance the sinterability of the porous SiC ceramics.

#### 3.2. Microstructure

Large spherical pores derived from spherical PMMA pore formers were clearly observed by SEM. In the porous SiC ceramics with B-C additives, a slight amount of platelike SiC grains was observed but most of SiC grains were not grown and they were fine. On the other hand, Al addition with B-C system enhanced grain growth of SiC and a large amount of plate-like SiC grains was formed. Notably, the porous SiC ceramics sintered with 0.1 wt% Al additive consisted of plate-like SiC grains of around 10-20  $\mu$ m, and the plate-like SiC grains stuck out of the walls of pores and the sintered surface in the porous SiC ceramics. SEM observation indicated that the porous SiC ceramics with unique morphology of entangled large plate-like SiC grains were successfully achieved.

Figure 2 shows pore size distribution of the porous SiC ceramics sintered with 0.1 wt% Al and B-C additives. The pore size distribution of the porous SiC ceramics was bimodal with peaks around 1  $\mu$ m and 8  $\mu$ m. In consideration of SEM observation, pores with the size of around 1  $\mu$ m and 8  $\mu$ m were considered to be formed by the formation of gaps among plate-like SiC grains and by pore formers, respectively.

The fractions of SiC polytypes in the porous SiC ceramics sintered with 0.1 wt% Al and B-C additives was determined by the Ruska's method based on XRD patterns. For comparison, the fractions of SiC polytypes in the



Fig. 2 Pore size distribution of porous SiC ceramics sintered with 0.1 wt% Al and B-C additives

porous SiC ceramics without Al-B-C additives were also analyzed. The porous SiC ceramics without Al-B-C additives mainly consisted of 3C ( $\beta$ -SiC) and the fraction of 3C was 74 %. The fractions of 4H( $\alpha$ -SiC) and 6H( $\alpha$ -SiC) in the porous SiC ceramics without Al-B-C additives were 2 % and 15 %, respectively. In the porous SiC ceramics sintered with 0.1 wt% Al and B-C additives, the fraction of 3C significantly decreased from 74 % to 13 %, and the formation of 6H was enhanced from 15 % to 76 %. In addition, the fraction of 4H also increased from 2 % to 8 %. From this result, it is suggested that the plate-like SiC grains formed in the porous SiC ceramics sintered with 0.1 wt% Al and B-C additives mainly corresponded to 6H phase.

#### 3.3. Compressive strength

Figure 3 shows compressive strength of the porous SiC ceramics sintered with B-C and Al-B-C additives at room temperature. Compressive strength of the porous SiC ceramics with B-C additives was around 40 MPa. The porous SiC ceramics sintered with Al-B-C additives showed the highest compressive strength at 0.1 wt% Al additive and the value was 50 MPa. However compressive strength of the porous SiC ceramics sintered with more than 0.1 wt%Al additive decreased to around 10 MPa. These results seem to be derived from large grain growth in the porous SiC ceramics sintered with Al-B-C additives. Further studies such as fracture surface observation after compressive test have been ongoing to clarify the effect of Al-B-C additives on compressive strength of the porous SiC ceramics.

#### 3.4. Thermal conductivity

Figure 4 shows thermal conductivity of the porous SiC ceramics sintered with B-C and Al-B-C additives at room temperature. The porous SiC ceramics sintered with B-C additives had thermal conductivity of 14 W/m•K, and thermal conductivity of the porous SiC ceramics sintered with Al-B-C additives ranged from 4 to 16 W/m•K. These values almost agreed with the change in bulk density, but



Fig. 3 Compressive strength of porous SiC ceramics sintered with B-C (Al : 0 wt%) and Al-B-C (Al: 0.03-2.0 wt%) additives at room temperature



Fig. 4 Thermal conductivity of porous SiC ceramics sintered with B-C (Al : 0 wt%) and Al-B-C (Al: 0.03-2.0 wt%) additives at room temperature

grain growth and solid solution of Al into SiC would affect their thermal conductivity. As grain size in the porous SiC ceramics becomes larger, their thermal conductivity would increase. It has been experimentally demonstrated that thermal conductivity of polycrystalline ceramics increases with their average grain size. As Al is doped into SiC crystal structure of the porous SiC ceramics, their thermal conductivity would decrease. It has been reported that solid solution of impurities into ceramics decreases their thermal conductivity. It is necessary to investigate the effects of grain growth and solid solution of Al into SiC crystal structure on thermal conductivity of the porous SiC ceramics in order to achieve higher thermal conductivity.

#### 4. Conclusion

In this study, porous SiC ceramics with in-situ platelike grain growth were fabricated by pressureless-sintering using Al, B and C additives, and the effects of Al, B and C additives on the microstructure of porous SiC ceramics and their properties were investigated. Bulk density of the porous SiC ceramics sintered with Al-B-C additives ranged from 1.44 to 1.92g/cm<sup>3</sup>, and their open porosity varied from 37 to 53 %. Whereas the porous SiC ceramics sintered with B-C additives mostly consisted of fine SiC grains without grain growth, Al addition with B-C system enhanced platelike grain growth of SiC in the porous SiC ceramics. Notably, the porous SiC ceramics sintered with 0.1 wt% Al and B-C additives consisted of plate-like SiC grains of around 10-20 µm, and the plate-like SiC grains stuck out of the walls of pores and the sintered surface in the porous SiC ceramics. The pore size distribution of the porous SiC ceramics was bimodal with peaks around 1 µm and 8 µm, and pores with the size of around 1 µm and 8 µm were considered to be formed by the formation of gaps among plate-like SiC grains and by pore formers, respectively. XRD results suggested that the plate-like SiC grains formed in the porous SiC ceramics sintered with Al-B-C additives during sintering mainly corresponded to 6H ( $\alpha$ -SiC) phase. The porous SiC ceramics sintered with Al-B-C additives showed highest compressive strength at 0.1 wt% Al additive and the value was 50 MPa. However compressive strength of the porous SiC ceramics sintered with more than 0.1 wt% Al additive decreased to around 10 MPa, and these results seem to be derived from large grain growth in the porous SiC ceramics sintered with Al-B-C additives. Thermal conductivity of the porous SiC ceramics sintered with Al-B-C additives ranged from 4 to 16 W/m•K, and these values almost agreed with the change in bulk density, but grain growth and solid solution of Al into SiC would affect their thermal conductivity.

From these results, it is concluded that high performance porous SiC ceramics with unique morphology of entangled large plate-like SiC grains were successfully achieved based on in-situ grain growth.

#### Acknowledgment

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D. Advanced Medical Application Division

## D.1 PIXE Analysis of Cs in Vegetables Grown by Hydroponics

Yoshiyuki OGURI and Wu WEIQING

#### 1. Introduction

Experimental data on the absorption of Cs by vegetables from the culture environment are important for assessing internal exposure due to radioactive Cs isotopes. Hydroponic culture is a suitable technique for basic experiments, because the culture environment is much simpler than that of soil-based methods, and can be precisely controlled.

In this note we investigate the applicability of PIXE (Particle Induced X-ray Emission) analysis to the measurement of Cs in vegetables cultured by hydroponics.

#### 2. Experimental

Details of the experiment has been reported elsewhere[1]. As the sample vegetable, pea (*pisum sativum L*.) was cultivated from seeds using a commercially available hydroponic grow box (Green Farm Cube UH-CB01G1, U-ING) for two weeks. The culture solution was prepared by adding different amounts of  $Cs(NO_3)_2$  to the original solution containing B, N, Na, Mg, P, K, Ca and Mn. After harvesting, the plants were cut into leaf, stem, root and seed sections. Each part was incinerated and the ash was dissolved in water containing Y as an internal standard. 10-µL of the ash solution was dropped on a 6-µm thick polypropylene foil and dried in room temperature.

These target samples were irradiated with a 2.5-MeV proton beam with a beam current of 1 nA. The emitted X-rays were detected by a 30-mm<sup>2</sup> Si(Li) detector. A 50-µm thick mylar foil was placed in front of the detector to reduce the low-energy background X-rays. The measuring time was 10 min for each sample.

#### 2. Results and Discussion

Measured PIXE spectra for the leaf section, as an example, are shown in Fig. 1. We see Cs-LX-ray peaks for the sample grown by the culture solution with Cs. All other elements with Z > 11 in the solution were also identified.

Figure 2(a) and (b) respectively show the Cs and K concentrations in the plant as a function of the Cs concentration of the culture solution. The Cs concentration in the plant increased with increasing the Cs concentration in the solution. We found that, particularly at high Cs concentrations, accumulation to the leaf is stronger than those in other parts. On the other hand, as seen in Fig. 2(b), the K concentration does not clearly change with Cs concentration in the solution.

#### 3. Conclusions

The limit of detection for Cs was not lower than 100 ppm, mainly because the large Ca-K<sub> $\beta$ </sub> peak (4.01 keV) interferes with the small Cs-L<sub> $\alpha$ </sub> peak (4.29 keV) in the PIXE spectrum. Nevertheless, we found that the analytical

method based on PIXE is applicable to experimental study on the Cs absorption by vegetables grown by hydroponic culture, if the Cs concentration is enough.

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Fig. 1 PIXE spectra for the leaf section of peas cultivated by the solution (a) with and (b) without Cs.



Fig. 2 (a) Cs and (b) K concentration in each section as a function of Cs concentration in the culture solution.

### D.2 Development of human skin 3D organoid for evaluation of radiation effect

Mikio SHIMADA, and Tomoko MIYAKE

#### 1. Introduction

Ionizing radiation (IR) causes genomic instability and chromosome aberration leading to tumorigenesis in the cellular level. Estimating the biological effect and cancer risk of IR to each organ is important for safe and development of radiation protection method. Skin tissue is an external organ in the human body and plays several roles in maintaining body metabolism, such as skin respiration, sweating, and protection from dryness and bacterial infection. The epidermis is the outermost layer of the skin and usually has been exposed to genotoxic stresses, such as IR, ultraviolet (UV) rays, and chemical compounds. These factors attack the genomic DNA and cause severe DNA damage, leading to genetic mutation and tumorigenesis. However, the evidence to evaluate radiation effect of skin tissue are limited because of difficulty of experimental technique in vivo and in vitro. In this study, we developed three dimensional (3D) organoid experimental model using skin keratinocytes derived from human induced pluripotent stem cells (iPSCs). We assessed DNA damage and cellular senescence in 3D skin organoid after IR. Our 3D organoid model will contribute to elucidate skin reaction after IR.

#### 2. Materials and Methods

#### 2.1. Differentiation of iPSCs into keratinocytes

For 1 day,  $8 \times 10^4$  single cells of iPSCs were cultured in Nutristem containing 5 µM Y27632 (WAKO, Japan) on iMatrix-511 coated 10 cm dish. The iPSCs were then incubated in DKSFM (Thermo Fisher Scientific) supplemented with 1 µM RA (Sigma-Aldrich) and 10 ng/mL BMP4 (R&D Systems, USA) for 4 days. And then the medium was replaced with DKSFM supplemented with 20 ng/mL EGF (R&D Systems) and cultured for 10 days, at which time the first passage was performed. iPSC-derived keratinocytes were seeded at 3  $\times$  10<sup>4</sup> cells/cm<sup>2</sup> and cultured in DKSFM supplemented with 10  $\mu M$  Y27632 and 20 ng/mL EGF on 0.03 mg/mL type I collagen (Advanced BioMatrix, USA) and 0.01 mg/mL fibronectin (Sigma-Aldrich) coated dish. On days 24 and 34, the second and third passages were performed using the same protocol use for the first passage.

#### 2.2. Construction of 3D skin organoid

 $2 \times 10^4$  NB1RGB were embedded in type I collagen gel (Nitta gelatin) in the 0.4 µm pores insert (Thermo Fisher Scientific) attached to a 24 well plate, and cultured in DMEM for 1 week. Then 15 ×10<sup>4</sup> keratinocytes (KCs 1stP) differentiated from iPSCs were seeded on collagen gel and cultured for 2 days in DKSFM supplemented with 20 ng/mL EGF and 10  $\mu$ M Y27632. Primary keratinocytes (HEKn) were similarly cultured in EpiLife containing 60  $\mu$ M calcium chloride. And then, the insert was transferred to 6 cm dish, the medium inside of insert was removed, and the keratinocyte layer was exposed to the air (air lift). The medium (DMEM: KSFM (Thermo Fisher Scientific) = 1: 1, adjusted to 1.8 mM calcium chloride) was placed outside of the insert. The medium was changed every 2 or 3 days and cultured for 14 days.



Fig. 1 2<sup>nd</sup> passage keratinocytes derived from hiPSCs. Cells were stained keratinocytes marker K14 and p63 antibodies, pluripotent stem cells (PSCs) marker OCT4 antibody. 2<sup>nd</sup> passage keratinocytes were positive to the keratinocytes marker but not PSCs maker.

#### 3. Results and Discussion

#### 3.1. Generation of keratinocytes from iPSCs

We first attempted to derive keratinocytes from human iPSCs. We used iPSCs generated from human neonatal fibroblasts NB1RGB. Hereafter, we used skin differentiated cells on Days 21, 31, and 41 as first passage (P1) keratinocytes, second passage (P2) keratinocytes, and third passage (P3) keratinocytes, respectively. All these cells expressed keratinocyte markers, namely keratin 14 (K14), epidermal basal keratinocyte expression protein, and p63, a p53 homolog transcription factor (Figure 1). Octamer-binding transcription factor 4 (OCT4), which was used as a Pluripotent stem cells (PSCs) marker, was undetectable in derived keratinocytes, confirming the differentiation from iPSCs to keratinocytes (Figure 1). Collectively, the cell morphology and staining of keratinocyte markers showed that derived cells have a

keratinocyte-like character.



Fig. 2 3D skin organoid model. (A) Passage 1 keratinocytes derived from hiPSCs and primary skin keratinocytes were used for 3D organoid. Cryosection of skin tissue was immune-stained with keratinocytes marker K14 and K10 antibodies. (B) After IR exposure, Cryosection of 3D organoid was immune-stained with keratinocyte marker K14 antibody and DNA double strand breaks marker 53BP1 antibody.

#### 3.2. Skin function and DDR in 3D skin organoid

To analyze skin function, we established 3D organoid culture using iPSCs derived keratinocytes. For the construction of 3D organoid, P1 keratinocytes were layered on skin fibroblasts. Primary keratinocytes HEKn were used for control cells. 3D skin layer structure was confirmed with K14 antibody for basal cell layer and K10 antibody for suprabasal cell layer (Fig 2). Furthermore, iPSCs derived keratinocytes and HEKn 3D organoid structure showed 53BP1 foci after IR irradiation (Figure 2). These data suggested that this 3D skin organoid model is useful tool to asses molecular mechanism such as DNA damage response and apoptosis after IR.

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# D.3 Investigation of mass-separated particle flux from a laser-ablation aluminum cluster source

Jun HASEGAWA

#### 1. Introduction

When an energetic cluster ion is injected into solid material, a coincident impingement of multiple atoms composing the cluster induces unique phenomena such as crater formation, an enhancement of sputtering yield, and an increase of stopping power. Although a lot of studies on the irradiation effect of high-energy cluster ions have so far been conducted, beam energies per nucleon were typically limited to less than 1 MeV/u because of the upper limit of the acceleration voltage of electrostatic accelerators. Takayama et al. have recently proposed the concept of the induction microtron, which allows us to accelerate large cluster ions (~1000 nucleons) from 1 keV/u to more than 1 MeV/u.

The induction microtron requires injection of a pulsed cluster ion beam with a pulse duration of typically a few microsecond. The laser-ablation cluster ion source is considered to have the potential to supply a high flux cluster beam to the microtron because it adopts a combination of laser ablation and supersonic expansion of high pressure buffer gas to produce of dense metal vapor containing a large amount of clusters having high directionality. Although many studies employed the laser-ablation cluster sources to investigate physical properties and chemical reactions of metal clusters, correlations between operational parameters of the cluster source and the available flux of cluster particles have not so far been studied sufficiently.

The purpose of this study is to explore how intense cluster beams can be obtained actually by the laser-ablation cluster source. The amount of cluster particles depends on the clustering process, which are determined by state quantities such as density and temperature of the metal vapor. In addition, the transportation process, in which the ablation vapor stagnates in a waiting room and then ejects with a supersonic helium gas flow through a nozzle, is considered to affect finally available cluster fluxes. Thus, we investigated flux waveforms of aluminum atoms and clusters supplied from a laser-ablation cluster source under various operational conditions (backing helium pressure, laser fluence, and exit hole diameter of a conical nozzle), and discussed how these parameters affect the clustering and transportation processes.

#### 2. Experimental Setup

Figure 1 shows the geometry of the cluster source developed in this study. A frequency-doubled pulsed Nd:YAG laser irradiated an aluminum disk target ( $\phi$ 25 mm) mounted on a motorized rotational stage. The rotation speed of the stage was set to ~20 rpm to obtain good reproducibility of laser ablation. Pulsed helium gas was

supplied through a fast valve (open time ~1 ms) with a backing pressure ( $P_{\text{He}}$ ) ranging from 0.3-1 MPa. To perform laser ablation after helium gas completely filled a waiting room, the time interval between the onset of valve opening and laser irradiation was set to 700 µs. The laser spot area on the target surface was evaluated to be ~0.2 mm<sup>2</sup>. Metal vapor produced by laser irradiation was carried into vacuum through a conical nozzle by a strong helium flow. Two conical nozzles with two different exit-hole diameters ( $\phi$ 1.5 mm and  $\phi$ 2 mm) were compared to examine the effect of the gas flow speed on the generation and transportation of the clusters.

The whole experimental setup is shown in Fig. 2. A voltage of 500 V was applied to deflection plates (A) at the exit of the cluster source to eliminate charged particles and observe only neutral particles downstream. An array of skimmers and a  $\phi$ 3-mm aperture collimates the cluster beam and a two-stage differential pumping keeps the pressure in the analysis region less than 10<sup>-4</sup> Pa. A time of flight mass spectrometer (TOFMS) was located 65 cm downstream from the laser target. A KrF excimer laser



Fig. 1. The cross-sectional view of the cluster source.



Fig. 2. A schematic of the experimental setup.

illuminated a 1 mm×1 mm area in the acceleration gap of the TOFMS, and ionized neutral clusters. The interval between the ablation laser and the ionization laser ( $\tau_D$ ) was precisely controlled by a pulse delay generator. Ionized clusters were orthogonally accelerated by a two-stage acceleration gap (20 kV in total) and finally focused onto a micro channel plate (MCP) by an einzel lens. To observe relatively small signals of clusters, a deflection voltage of 400 V was applied to deflection plates (B) behind the acceleration gap to prevent small particles such as Al, AlO, Al<sub>2</sub>, and Al<sub>2</sub>O that have much higher signal intensities than clusters from reaching the detector and disturbing the sensitive measurement of cluster signals. Although the deflection field modifies also the trajectories of large cluster ions, those cluster ions (~ $10 \le N \le ~100$ ) could be detected with high efficiency (~100%) by tuning the focusing field of the einzel lens. The MCP signal was averaged over 64-512 shots to reduce statistical errors and increase signal to noise ratios.

#### 3. Results and Discussion

Figure 3 shows a typical TOFMS spectrum observed with a laser fluence of 2 J/cm<sup>2</sup>. The inserted figure shows a spectrum of large clusters (N > 5), which was obtained with a deflection voltage. As shown in the figure, the signal intensities of small particles were much larger than those of large clusters. As shown in the figure, the size distribution has a local maximum around N = 25. An increase of ionization laser fluence enhanced the abundance of all particles and slightly changed the cluster size distribution; the relative abundance of small clusters ( $N \sim 14$ ) increased and that of large clusters ( $N \sim 35$ ) decreased with increasing laser fluence. We consider that these changes were caused by the photodissociation of large clusters.

The flux waveform of each kind of particle was also extracted from the TOFMS signals by examining the temporal change of the intensity of the corresponding mass peak. Figure 4 shows the reconstructed flux waveforms of aluminum atoms and clusters (N = 7, 14, 25, 35, and 50). The arrival time of aluminum atoms ranged from 400 to 700 µs and its waveform had a sharp peak at 500 µs. Flux waveforms of the clusters had similar features (arrival time and maximum time) to that of the aluminum atoms. The speed of these particles (atoms and clusters) is estimated to



Fig. 3. Typical mass spectra obtained by the TOFMS.



Fig. 4. Reconstructed flux waveforms of (a) aluminum atoms (b) cluster particles observed with  $P_{\text{He}} = 0.9 \text{ MPa}$  and  $I_{\text{L}} = 2 \text{ J/cm}^2$ .



Fig. 5. Backing pressure dependences of (a) Al atom and (b) Al cluster beam fluxes.

be 1600 m/s from the time of flight ( $\sim$ 400 µs) from the cluster source to the ionization point (65 cm).

Backing helium pressure dependences of the aluminum atom flux and the cluster fluxes are shown in Fig. 5(a) and (b), respectively. When the pressure was relatively low ( $P_{\text{He}}$ )  $\sim 0.3$  MPa), the aluminum flux was very small and the arrival time of aluminum atoms ranged from 500 to 700 µs. Meanwhile, clusters were not observed with this pressure. The aluminum atom flux dramatically increased and its arrival time became earlier (400-700  $\mu$ s) when the backing pressure increased to 0.5 MPa, but they changed not so much with  $P_{\text{He}} > 0.5$  MPa. This increase in the aluminum atom flux suggests that the diffusion of the ablation vapor plume was strongly suppressed by high pressure background helium gas in the waiting room. On the other hand, cluster signal intensities were lower than noise levels with  $P_{\text{He}} = 0.3$  MPa and Al<sub>14</sub>, which had relatively large abundance, could be observed with backing pressures more than 0.5 MPa. The flux of  $Al_{14}$  had a maximum at 520  $\mu$ s after the aluminum atom flux had a maximum at 460 µs. The cluster signal intensity observed with  $P_{\text{He}} = 0.9$  MPa was twice higher than that with 0.7 MPa, showing that cluster generation was enhanced by increasing backing



Fig. 6. (a) Flux waveforms of aluminum atoms and clusters (N = 35) observed with the  $\phi$ 2-mm nozzle with  $P_{\text{He}} = 0.9$  MPa. (b) Flux waveforms of aluminum atoms and clusters (N = 35) with the  $\phi$ 1.5-mm nozzle with cluster fluxes (N = 14 and 35). Laser fluence was 2 J/cm<sup>2</sup> in both cases

pressure.

Figures 6(a) and (b) show the aluminum atom and cluster flux waveforms observed with conical nozzles having two different exit-hole diameters ( $\phi$ 2 mm and  $\phi$ 1.5 mm). When we used the  $\phi$ 2-mm nozzle, the aluminum atom and cluster fluxes reached maximum values at  $I_{\rm L} = 2 \, \text{J/cm}^2$  and decreased with increasing laser fluence more than 3 J/cm<sup>2</sup>. This is probably because plasma production (ionization) in the ablation plume became more remarkable with higher laser intensities. On the other hand, when the  $\phi$ 1.5-mm nozzle was employed, the aluminum atom and cluster fluxes became reached maximum values with relatively low laser intensity (~1 J/cm<sup>2</sup>) compared with the case using the maximizing the aluminum atom flux is probably due to the difference in the helium pressure in the waiting room. The flow rate of the helium gas was limited by the inlet of the waiting room because the diameter of the inlet (1 mm) is smaller than the exit hole diameter. So the pressure in the waiting room can be higher with  $\phi$ 1.5-mm nozzle than with  $\phi$ 2-mm nozzle even if the backing pressures are the same. The arrival time was delayed by 440-800 µs when the smaller ( $\phi$ 1.5 mm) exit hole was used, indicating that the helium gas flow in the waiting room was slowed down. The arrival time of the clusters (N = 35) showed the same trend as the atoms. The flux waveforms of the clusters were also similar to those of the aluminum atoms.

We compared the aluminum cluster yield (N = 35) with the aluminum atom yield by integrating the flux waveforms in time to evaluate the degree of clustering. The cluster



Fig. 7. Aluminum cluster yield (N=35) versus aluminum atom yield. The fitting line represents a power function. The index of the function is 2.

yield nonlinearly increased with respect to the atom yield as shown in Fig. 7. This quadratic relation may result from three body recombination, whose reaction probability is proportional to the square of the aluminum atom density. An enhancement of the aluminum atom density is expected to enhance cluster generation. Increasing laser energy with the same laser fluence can be a way to obtain larger cluster flux because the laser fluence has optimum values for cluster generation (see Fig. 6).

#### 4. Conclusion

To design high flux cluster beam sources required for high-energy circular cluster accelerators, we conducted an experimental survey on how the operational parameters of laser-ablation cluster source affect the cluster size distribution and flux waveforms. We observed aluminum clusters with sizes up to 100. Bunch lengths of the cluster beams were typically around 300 µs. The speed of clusters was estimated to be about 1600 m/s, which is almost corresponding to the speed of the supersonic helium gas flow. This result shows that the helium gas acted as a carrier of the clusters and also a cooling media of the aluminum vapor. Backing pressures beyond 0.5 MPa were found to be necessary to generate and carry clusters efficiently. Also, we found that there were optimum laser fluences (1-2 J/cm<sup>2</sup>) for aluminum cluster generation. The cluster yield increased quadratically with respect to the atom yield, which suggests that increasing laser pulse energy while maintaining the optimum laser fluence will drastically improve cluster generation efficiency.

#### Acknowledgment

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E. Fundamental Research Division

## E.1

## Study for nuclear fission and its application

Chikako ISHIZUKA and Satoshi CHIBA

#### 1. Introduction

Nuclear fission has been studied for eighty years since its discovery. However, we still cannot explain the whole property of nuclear fission even of <sup>236</sup>U with sufficient predictive power, although empirical models have been applied to practical use. Such practical models may work well in the case of well examined n-induce fission of <sup>235</sup>U and <sup>239</sup>Pu. On the other hand, in order to develop a new system such as Accelerator-driven Systems (ADS) and Fast Reactors (FR), and in order to reduce the TRU wastes, we need high quality data on minor actinides (MA) as well as n-induced <sup>235</sup>U fissions. Experiments to obtain fission data for MA and long-lived fission products (LLFP) have been performed in various facilities. But it is still difficult to cover whole fission data, such as fission fragment mass distributions (FFMDs), thermal energy (total kinetic energy; TKE), prompt and delayed neutrons, and decay heat from the fission products. Chiba laboratory has developed nuclear models which can reproduce and predict these physical quantities on nuclear fissions.

In addition to the above fundamental approaches, we are also working on the reduction of poisonous nuclear wastes by studying the whole mechanism of nuclear fission process from scission to  $\beta$ -decay, and by developing high-quality nuclear data. In our laboratory, we have also



Fig.1 Schematic view of the whole process of nuclear fission.

investigated the influence of nuclear data on the decommissioning costs. In Fig.1, we show the schematic view of nuclear fission process. The aim of Chiba laboratory is the improvement of fission nuclear data such as fission product yields, prompt neutrons and decay heats, by eludidating the fundamental mechanism of nuclear fission.

#### 2. Fundamental studies for nuclear fission

Nuclear fission is the motion of a many-body quantum system. It is still very difficult to describe the whole feature of a nuclear fission process with a single model. Indeed, there is no theoretical model, which can completely simulate the whole process of nuclear fission shown in Fig.1. Therefore, we have adopted different models depending on our purpose. In our laboratory, we have studied the fundamental mechanism of nuclear fission at low energies using various nuclear-physics theories such as multidimensional Langevin models, Anti-symmetrized Molecular Dynamics model, and Time-dependent Hartree Fock model.

#### 2.1. Multidimensional Langevin models

Langevin models can reproduce and predict not only the fission product mass yields but also the total kinetic energies of the fission fragments of actinides. In the Lanvin model, a nuclear fission process is regarded as a time-evolution of the nuclear shape of a fissioning nucleus following the equation of motion under the friction force and the random force. Such equation of motion is called the Langevin equation. We have developed the Langevin model by extending degree of freedom to describe a realistic fission-nuclear-shape, and by introduction of the microscopic transport coefficients based on the linear response theory. As a result, we found that correlated transition of the total kinetic energy and the mass distributions of fission fragments [1]. We also found that temperature dependence of the shell effect plays an essential role to explain fission fragment mass distributions of Th-isotopes [2].

#### 2.2. Anti-symmetrized Molecular Dynamics (AMD) model

The Langevin models cannot directly provide the spins of fission fragments and the division of nuclear excitation energy among fission fragments. Those quantities have been investigated by the Quantum molecular dynamics (QMD). However, a nucleus is a quantum many-body system of fermions such as protons and neutrons. Fermions are anti-symmetric under particle exchange. Considering such characteristics of the nuclear many-body system correctly, we need to extend the QMD to include such anti-symmetrization for more precise description of nuclear fissions. The extended model is called the Anti-symmetrized Molecular Dynamics (AMD), which is a very powerful model to dee scribe both nuclear reactions and nuclear microscopic study of nuclear fission mechanisms and applied it to the <sup>236</sup>U fission for the first time. We also applied the AMD to investigate the detailed fission mechanisms such as angular momentum, which are difficult to elucidate using the Langevin model.

#### 2.3. Time-dependent Hartree Fock (TDHF) model

In the standard Langevin models, the friction force is provided by the potential derivatives with a macroscopic coefficient evaluated the one-body dissipation model. In order to valid such friction force, we have performed the Time-dependent Hartree Fock (TDHF) calculations. The TDHF model can derive the friction coefficient considering all nucleon degree of freedom. Then we study the friction coefficient using TDHF to valid the friction strength we have used in the Langevin model.

#### 3. Beta decay of fission products

After prompt neutrons and gammas emitted from the fission fragments, the beta-decays of these nuclei will occur. Neutrinos produced by the beta-decay process play a significant role in the surveillance and in-service inspection in nuclear power plants. In our laboratory, we have studied the anti-neutrino spectrum from aggregate fission products beta-decays based on the gross theory.

#### 4. Evaluation of nuclear data

4.1: Development of nuclear fission data library for various applications

High precision nuclear data is necessary to economically evaluate the total heat from the fission products and their toxicity. Major nuclear data libraries such as JENDL-4.0 contain ambiguity due to the experimental data and human errors during coding process. In our laboratory, we have investigated the suspicious nuclear data, and have examined its influence on transmutation and decommissioning. For example, we found the possibility of 20% overestimation in the cross section of thermal neutron caption on <sup>102</sup>Ru of JENDL-4.0, where Ru-isotopes affect the transmutation efficiency using nuclear reactors.

We have also developed the Hauser-Fechbach model to predict the fission product yields and the isomar ratio [3]. Nuclear data is the most basic data for nuclear reactors. We have to pay much attention to its precision, and develop better nuclear data. For this aim, we have studied nuclear fission from the fundamental mechanism.

#### 4.2: Estimation of uncertainty of nuclear data

Many nuclide data in JENDL-4.0 does not have uncertainty which is important for quantitative estimation of many quantities in radiator physics. We have evaluate the uncertainty of the cross sections of some LLFP nuclides in JENDL-4.0 by use of T6 code which evaluate the nuclear data employing the Bayesian Monte Carlo calculations. Using these results, we have also evaluate the uncertainty of the nuclear transmutation of LLFP nuclides, and so on.

#### Acknowledgment

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## Possibility of OES Measurement of Electron Energy Distribution Function of Atmospheric-Pressure Non-Equilibrium Plasma

Hiroshi AKATSUKA and Atsushi NEZU

#### 1. Introduction

Atmospheric-pressure non-equilibrium plasmas have been widely applied in many fields. However, it is still difficult for us to measure electron temperature and density of these plasmas, to say nothing about the electron energy distribution functions (EEDF). On the other hand, in this plasma, continuum emission is generated due to the bremsstrahlung of free electrons in the plasma against neutral particles with sufficient intensity in the near UV through visible wavelength range. The objective of this study is to consider the spectral dependence of the continuum emission of the atmospheric-pressure nonequilibrium plasmas on the EEDF.

#### 2. Experiments

Figure 1 shows the schematic diagram of the discharge apparatus, whose further details were described elsewhere [1]. The 50-Hz AC discharge power was supplied by a stepup transformer with its maximum voltage  $\sim 9$  kVp-p.

The spectroscopic characteristics of the plasma can be observed through the optical window equipped with the discharge tube. Figure 2 shows the OES result observed experimentally as a time average over a discharge period. By theoretical fitting procedure as explained later, we can determine the electron temperature for the Maxwellian or Druyvesteynian EEDF as the best fitting parameter [2, 3].

#### 3. Discussion

The continuum spectrum emitted from this kind of plasma is attributed to the electron-atom (e-a) bremsstrahlung in the plasma, which is theoretically expressed by the following equation [1]:

$$\varepsilon_{ea} = \sqrt{\frac{2}{m_e}} \frac{N_e N_a}{\lambda^2} \frac{hc}{4\pi} \int_{hv}^{a} Q_{ea}^{B}(\lambda, \varepsilon) \sqrt{\varepsilon} f(\varepsilon) d\varepsilon, \qquad (1)$$

where  $N_e$  is the electron density,  $N_a$  is the atom density,  $\lambda$  is



Fig. 1 Schematic diagram of the discharge tube. The electrodes are immersed in water in doubly sealed quartz tube. The outer tube is filled with the discharge rare gas.



Fig. 2 Spectrum observed from the atmospheric-pressure nonequilibrium argon plasma with (a) the Maxwellian EEDF and (b) the Druyvesteynian EEDF. In (b),  $T_e$  is defined to be  $kT_e \equiv$ (2/3) $< \epsilon >$ , where  $< \epsilon >$  is the electron mean energy and k is the Boltzmann constant.

the wavelength of the continuum emission, *h* is the Planck constant, *c* is the speed of light,  $\varepsilon$  is the electron energy, *v* is the frequency of the emitted photon ( $v = c/\lambda$ ),  $f(\varepsilon)$  is the EEDF, and  $Q_{ea}^{B}$  is described with the momentum transfer cross section  $Q_{ea}^{mom}$  as

$$Q_{ea}^{B}(\lambda,\varepsilon) = \frac{8\alpha}{3\pi} \frac{\varepsilon}{m_{e}c^{2}} \left(1 - \frac{hc}{2\lambda\varepsilon}\right) \sqrt{1 - \frac{hc}{\lambda\varepsilon}} Q_{ea}^{mon}(\varepsilon), \qquad (2)$$

where  $\alpha$  is the fine structure constant.

Equations (1) - (2) enable us to calculate the continuum spectrum emitted from the plasma as the bremsstrahlung, when we assume the EEDF. In Fig. 2, we added the theoretical emission spectra calculated with the Maxwellian EEDF in (a) and Druyvesteyn in (b) [4-5].

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## **E.3**

## Measurement of Underwater Ar Arc Plasma by Optical Emission Spectroscopy

Hiroshi AKATSUKA, Atsushi NEZU and Shinsuke MORI

#### 1. Introduction

Establishing a method for decommissioning the Fukushima Daiichi Nuclear Power Stations is an urgent issue. One of the most important issues is the removal of nuclear fuel debris. It is necessary to cut and crush the lumped fuel debris into an appropriate size so as to be easily taken out, where a method of thermally performing by arc plasma is proposed.

It is desirable to cut, crush and take out fuel debris as much as possible in water, because shielding of radiation by water and cooling of fuel can be expected, and furthermore, scattering of fumes containing radioactive substances can be prevented. In order to realize safe decommissioning, this research is conducted for the purpose of measuring basic parameters and clarifying characteristics of underwater arc plasma.

#### 2. Experiments

Figure 1 shows a schematic of the arc discharge plasma generator and peripheral equipment used in this study. The emission of the plasma plume was measured by a spectrometer through a quartz glass window attached to the side of the chamber. The chamber was filled with water so that both electrodes were fully immersed. The flow rate of argon is 30 L/min. Since the chamber is open to the atmosphere, the internal pressure is equal to the atmospheric pressure.

The measurement object of this research is atmospheric pressure arc plasma. That is, it is considered that the assumption of local thermal equilibrium is sufficiently established. Hence, the observed excitation temperature  $T_{ex}$  is considered to be almost the same with the electron temperature, where emission lines of Ar I shown in Table 1 are applied. These lines can be observable within one frame of polychromator, and consequently, the simultaneous intensity of Ar I is determined.

On the other hand, electron density  $n_e$  is determined with the Stark broadening of Balmer- $\alpha$  line of H atom, which is found inevitably as a dissociated fragment of water molecule.



Fig. 1 Schematic of the experimental apparatus.

Table 1. Spectral lines observed in this study and some of their constants.

$\lambda_{pq}$ [nm]	$E_p$ [eV]	$A_{pq} [10^7 \text{ s}^{-1}]$	$g_{pq}$	Transition
687.13	14.71	0.278	3	4d - 4p
693.77	14.69	0.308	1	4d - 4p
696.54	13.32	0.0639	3	4p'-4s*
703.03	14.83	0.267	5	6s – 4p
706.72	13.30	0.380	5	4p'-4s*









#### 3. Results and discussion

From the Boltzmann plot, we obtain  $T_{ex}$  of the underwater arc plasmas as shown in Fig. 2.  $T_{ex}$  of gas-phase arc plasmas is obtained in the same way. They both increase as the current increases, however,  $T_{ex}$  of underwater ones is higher than those of gas-phase ones. It is considered that since specific heat capacity of H<sub>2</sub>O is larger than that of argon gas, thermal pinch effect works stronger for underwater ones.

Figure 3 shows  $n_e$  of each plasma. Similar to  $T_{ex}$ , it increases as the current increases in the order of  $10^{16}$  cm<sup>-3</sup> and  $n_e$  of underwater ones is higher than that of gas-phase ones. This is because in addition to contraction by thermal pinch effect, ionizing degree increases as the temperature gets higher, which is consistent with Saha's equation.

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## E.4 Diagnostics of Electron Temperature and Density of Low-pressure Microwave Discharge Ar Plasma by Optical Emission Spectroscopy Based on Collisional Radiative Model

Hiroshi AKATSUKA and Atsushi NEZU

#### 1. Introduction

Diagnostic methods of electron temperature  $T_e$  and density  $N_e$  that can be performed only by passive emission spectroscopy are not well established for non-equilibrium plasmas. Therefore, the development of a method that can diagnose  $T_e$  and  $N_e$  using a relatively simple and inexpensive device is desired. Therefore, a model that diagnoses  $T_e$  and  $N_e$  was created from multiple emission lines based on the Collisional Radiative Model (CR model) [1]. In this study, the usefulness of the created diagnostic model was examined experimentally.

#### 2. Model for the diagnostics

In this research, Ar plasma is targeted. The diagnostic model proposed in this study solves the conventional CR model in reverse. That is, it finds  $T_e$  and  $N_e$  by solving the rate equation in reverse using the excited state number density obtained from optical emission spectroscopy (OES) measurement as an input value. At that time, the Ar gas pressure was assumed to be  $1.33 \times 10^2$  Pa, the atomic temperature to be 500 K, and the electron energy distribution function (EEDF) was assumed to be Maxwell or Druyvesteyn distribution.  $T_e$  [eV] and  $N_e$  [cm<sup>-3</sup>] were assumed to be in the range of  $2.0 \le T_e \le 4.0$  and  $1.0 \times 10^{10}$  $\leq N_{\rm e} \leq 1.0 \times 10^{12}$ , respectively. However, in the case of the distribution, Druyvesteyn the effective electron temperature  $T_{\rm eff}$  was defined as the average energy  $< \varepsilon > =$ (3/2)  $k_{\rm B}T_{\rm eff}$  ( $k_{\rm B}$ : the Boltzmann constant). The model generated is the one in which  $T_e$  is diagnosed with 5p and 6p levels, and  $N_e$  is with the 4s, 4p and 5s levels.

#### 3. Experiments

Microwave discharge plasma was generated with 2.45 GHz microwave, with the discharge tube radius of 1.3 cm and discharge power of 100, 300 and 500 W. To verify the diagnostic model described in Section 2,  $T_e$  and  $N_e$  at distances of 10, 14, and 18 cm in the discharge tube axial direction from the waveguide center were measured by the following two methods. (1) OES measurement: the light emission from the plasma was received with a polychromator calibrated absolutely, and the number density of excited states determined. (2) Electrostatic probe measurement:  $T_e$  and  $N_e$  were determined using a symmetric double probe for comparison.

#### 4. Results and discussion

As shown in Figures 1 and 2, the results of  $T_e$  and  $N_e$  by probe measurement and those by OES measurement were consistent in both Maxwell and Druyvesteyn distributions.



Fig. 2 Diagnostics Result of N<sub>e</sub>.

 $T_{\rm e}$  agrees with the tendency that it once lowered as the distance from the center of the waveguide increases, and  $N_{\rm e}$  agrees with the tendency that it increases with increasing electric power.

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III. Co-operative Researches

#### III. Co-operative Researches

#### III.1 Co-operative Researches within Tokyo Institute of Technology

- Possible measurements of nuclear data using laser based neutron sources, Chikako Ishizuka, Satoshi Chiba
- (2) Study on prompt-neutron emission mechanism of nuclear fission based on a statistical model, Chikako Ishizuka, Shin Okumura, Satoshi Chiba
- (3) Development of quantitative evaluation method for charge polarization using Langevin model, Chikako Ishizuka, Satoshi Chiba
- (4) Measurement of Underwater Arc Plasma by Optical Emission Spectroscopy and its Heat Transfer to Liquid Phase Injected into Water, Professor Shinsuke Mori, School of Materials and Chemical Technology

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

- Study on numerical kinetic analysis method by integral kinetic model with delayed neutrons, Hiroki Takezawa, Tokyo City University
- (2) Solid oxide electrolysis cell development for CO<sub>2</sub> reduction, JSPS Grant-in-Aid for Scientific Research (B), 2019-2022.
- (3) Carbon Recycling Ironmaking System, Research Committee on Carbon Recycling Ironmaking System, 54<sup>th</sup> Committee on Ironmaking, Japan Society for the promotion of Science, 2019-2021.
- (4) Development of liquid breeder blanket of fusion reactors, National Institute for Fusion Science
- (5) Development of liquid surface divertor of fusion reactors, National Institute for Fusion Science
- (6) Development of Subcritical Water Washing System for Cleanup and Reuse of Contaminated Soil, and Volume-reduction of Radioactive Waste, The Environment Research and Technology Development Fund, 1-1805.
- (7) Study on Cs desorption from soil cay minerals by subcritical water containing metal ions, Grant-in-aid for Scientific research B, 18H03398.
- (8) Study of effective ion recognition and selective separation of f-series elements based on higher oxidation states of Americium, Grant-in-Aid for JSPS Overseas Research Fellowship, Japan Society for the Promotion of Science, 434.
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- (14) Survey of radioactive waste solution, Japan Atomic Energy Agency, KY30N057.
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- (18) Collaborator, Advanced Research and Education Program for Nuclear Decommissioning, MEXT, KH300001.
- (19) Study on Low-Cost Process of SiC/SiC Composites: Japan Aerospace Exploration Agency (JAXA)
- (20) Study on Properties of B<sub>4</sub>C Neutron-Absorbing Materials for Control Rods: Japan Atomic Energy Agency (JAEA)
- (21) Study on Neutron-Irradiation Resistance of Orientation-Controlled Ceramics: National Institute for Materials Science (NIMS)
- (22) Sinterability of SiC Ceramics with Al<sub>4</sub>SiC<sub>4</sub> Addition and Their Properties: National Institute for Materials Science (NIMS)
- (23) Research on Formation and Characterization of Oxide Nanopowder : Vinca Institute, University of Belgrade, Serbia
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- (30) Research on adsorbents of siloxane compounds for application under extreme environment: Japan Aerospace Exploration Agency (JAXA)
- (31) Astrobiology Experiments Based on MeV Ion Beams, Division of Materials Science and Chemical Engineering, Faculty of Engineering, Yokohama National University.
- (32) Development of Negative Ion Sources for Tandem Accelerators, Atomic Energy Research Laboratory, Tokyo City University.
- (33) Detection and Quantification of Actinides by X-ray Fluorescence Analysis for Radiation Emergency Medicine, National Institutes for Quantum and Radiological Science and Technology
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- (36) The control of radiation dependent DNA damage response molecular network, Akihiro Katom Tohoku Medical and Pharmaceutical University
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- (39) Possible measurements of nuclear data using laser based neutron sources, with Takehito Hayakawa (QST)
- (40) Study on prompt-neutron emission mechanism of nuclear fission based on a statistical model, with Toshihiko Kawano (LANL, USA)
- (41) Development of quantitative evaluation method for charge polarization using Langevin model, with Katsuhisa Nishio (JAEA)

- (42) "Research and development of an innovative transmutation system of LLFP by fast reactors" entrusted to the Tokyo Institute of Technology by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT), with Toshio Wakabayashi (Tohoku Univ.), Yoshiaki Tachi (JAEA), Naoyuki Takaki (Tokyo City Univ.)
- (43) "Development of prompt-neutron measurement in fission by surrogate reaction method and evaluation of neutron- energy spectra" entrusted to JAEA by MEXT, with Katsuhisa Nishio (JAEA).
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- (45) "The study on nuclei and neutron matter using finite-range three-body force" Naoyuki Itagaki (Kyoto Univ.)
- (46) "The study on the influence of nuclear fission on the rapid neutron capture process" Shinya Wanajo (AEI, Max Plank Institute), Yuichiro Sekiguchi (Toho Univ.)
- (47) Measurement of Vibrational and Rotational Temperature in Spark-Discharge Plasma by Optical Emission Spectroscopy for Innovative Automobile Engine Research, TOYOTA Central R&D Labs
- (48) Determination of Electron Density and its Temperature using Collisional Radiative (CR) Model in High-density Helicon Plasma for Electric Propulsion of Artificial Satellites, Tokyo University of Agriculture and Technology, Chubu University
- (49) Diagnostics of Low-Pressure Discharge Argon Plasma by Multi-Optical Emission Line Analysis Based on the Collisional-Radiative Model, Tokyo Metropolitan Industrial Technology Research Institute
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## **IV.** List of Publications

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- (5) Van Khanh Hoang, Jun Nishiyama, Toru Obara: Design concepts of small CANDLE reactor with melt-refining process; *Progress in Nuclear Energy*, Vol. 108, pp. 233-242 (2018).
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- (3) Y. Kato: [Invited Lecture] "Active Carbon Recycling Energy System (ACRES) for the Next Low-Carbon Society"; 5th International Workshop on Heat/Mass Transfer Advances for Energy Conservation and Pollution Control, August 14(13-16), 2019, Novosibirsk, Russia.
- (4) Alemberti, K. Tucek, T. Obara, M. Kondo, A. Moiseev, I.S. Hwang, C. Smith, Y. Wu, THE GENERATION-IV LEAD FAST REACTOR ACTIVITIES, pp. 1580-1587(English), October 2-5, Russia, Moscow (2018).
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- (6) Toru Tsunoura, Katsumi Yoshida, Toyohiko Yano, Takuya Aoki, Toshio Ogasawara: High temperature bending behavior of SiC/Si binary eutectic alloys; 42nd International Conference and Exposition on Advanced Ceramics and Composites, Florida, USA, January 25, 2018, ICACC-S1-058-2018.
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- (11) Ryosuke S. S. Maki, Muhammad Fajar, Jelena Maletaškić, Anna Gubarevich, Katsumi Yoshida, Toyohiko Yano, Tohru S. Suzuki, Tetsuo Uchikoshi: Fablication of dense B<sub>4</sub>C/CNT composites and their thermal shock resistance; The 6th International Symposium on Advanced Ceramics (ISAC-6), Sendai, Japan, March 13, 2018, A13-10.
- (12) Jelena Maletaškić, Katsumi Yoshida, Toyohiko Yano, Jelena Luković, Jelena Zagorac, Branko Matović: High-Temperature Synthesis and Characterization of Boron Suboxide (B<sub>6</sub>O) and boron containing hard materials; The 6th International Symposium on Advanced Ceramics (ISAC-6), Sendai, Japan, March 13, 2018, C13-10.
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- (30) Y. Ishikawa, J. Hasegawa, K. Horioka: Mass separated particle flux from a laser-ablation metal cluster source; 22nd International Symposium on Heavy Ion Fusion and Beam-Driven High Energy Density Science, Daejeon, Korea, August 20-24, 2018, P-07.
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- Suzuko Saito, Jun Nishiyama, Toru Obara: Impact of Fuel and Coolant Material on Burnup Characteristics in Breed and Burn Fast Reactor; *Proc. of 2019 annual meeting of Atomic Energy Society of Japan*, 1K10 (2019).
- (2) Hiroki Osato, Jun Nishiyama, Toru Obara: Application of Melt-refining Process to Transition State of CANDLE Burning Fast Reactor; *Proc. of 2019 annual meeting of Atomic Energy Society of Japan*, 1K11 (2019).
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- (5) Y. Kato: [Invited Lecture] "Active Carbon Recycling Energy System for Low-Carbon Society"; 9<sup>th</sup> Symposium on Advanced Research of Energy and Materials, 2019/6/19, Sapporo.
- (6) Y. Kato: [Invited Lecture] Heat storage performance of high thermal conductivity composite using calcium hydroxide for thermochemical energy storage; 56th Japan Heat Transfer Symposium, OS-I311, 31 June, 2019, Tokushima.
- (7) S. Funayama, R.Guo, H. Takasu, Y. Kato: Heat storage performance of high thermal conductivity composite using calcium hydroxide for thermochemical energy storage; 56th Japan Heat Transfer Symposium, 29-31 June, 2019, Tokushima.
- (8) Rui GUO, Shigehiko FUNAYAMA, Hiroki TAKASU, Yukitaka KATO: Nano-modified materials for CaO/H<sub>2</sub>O/Ca(OH)<sub>2</sub> thermochemical energy storage; 56th Japan Heat Transfer Symposium, 29-31 June, 2019, Tokushima.
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- (10) Y. Maruyama, K. Nakajima, H. Takasu, Y. Kato: Development of solid oxide electrolysis cell for CO2 reduction in active carbon recycling energy system for iron-making process; *the Fifth International Symposium on Innovative Materials and Processes in Energy Systems* (IMPRES2019), P124, Poster presentation, 22(20-23) October, 2019, Kanazawa, Japan.
- (11) J. Kaneko, H. Takasu, K. Fujioka, Y. Kato: Development of thermochemical energy storage system by using metal chloride/ammonia system; the Fifth International Symposium on Innovative Materials and Processes in Energy Systems (IMPRES2019), P106, Poster presentation, 22(20-23) October, 2019, Kanazawa, Japan.
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- (25) Tomohiro Okamura, Eriko Minari, Masahiko Nakase, Hidekazu Asano, Kenji Takeshita: Evaluation of geological disposal area reduction index in various nuclear fuel cycle scenarios; Effects of Cs and Sr separation and horizontal emplacement configuration of waste package onreducing geological disposal area; 14<sup>th</sup> seminar on Reprocessing and Recycle Technology division, Atomic Energy Society of Japan, Tokyo, Japan, December 12, 2018.(Poster)
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- (33) Tatsuya Fukuda, Ryo Takahashi, Xiangbiao Yin, Yusuke Inaba, Miki Harigai, Masahiko Nakase, Kenji Takeshita: Reaction rate analysis of Cs removal reaction from contaminated soil using subcritical water treatment; 2019 Annual Meeting of Atomic Energy Society of Japan, Ibaraki, Japan, March 20-22, 2019.
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