## Effect of Soft X-rays on the Vacancy-type Defects in Highly Hydrogenated DLC Films

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**INTRODUCTION:** Diamond-like carbon (DLC) films are generally known to be highly resistant to X-ray irradiation. However, our group has reported that highly hydrogenated DLC (H-DLC) films with a hydrogen content greater than 40 at.% are modified by the irradiation. By the irradiation of soft X-rays to H-DLC film, the film volume and hydrogen content decrease and film hardness and  $sp^2/(sp^2 + sp^3)$  ratio of carbon atoms increase [1]. However, it is not known how the free volume in DLC films is changed by soft X-ray irradiation. In this study, we prepared samples with different doses of white soft X-ray irradiation and performed positron annihilation spectroscopy (PAS) measurements using the slow positron beam apparatus. We measured how vacancy-type defects in H-DLC films are changed by soft X-ray irradiation and discussed the hydrogen desorption process of H-DLC films by soft X-rays in terms of vacancy-type defects.

**EXPERIMENTS:** H-DLC film used in this work was deposited on Si wafers using an amplitude-modulated radio-frequency plasma-enhanced chemical vapor deposition method (Nippon ITF Co.). The irradiation of the H-DLC film by SR was carried out at BL06 of the NewSUBARU synchrotron facility. The SR at the BL06 sample stage had a continuous spectrum from infrared to soft X-rays, below 1 keV. In this work, four samples were prepared at SR dose, 500 mA h, 1000 mA h, 4000 mA h, and before irradiation. PAS measurements were performed using a slow positron beam system installed at the B-1 hole of the KUR. In the present study, we performed two kinds of PAS measurements, Doppler broadening and the positron annihilation lifetime spectroscopy.

**RESULTS:** The dependence of S-parameters and positron annihilation lifetimes (PAL) on soft X-ray dose is shown in Fig. 1. The positron lifetime increases with the soft X-ray dose. This means that the increase in density of H-DLC film due to soft X-ray irradiation is not due to a decrease in vacancy-type defect, but to a decrease in the composition ratio of hydrogen in the film.

On the other hand, the S-parameter was decreased by soft X-ray irradiation. In H-DLC films, the vacancy-type defect is considered to be almost entirely surrounded by hydrogen, which has only valence electron. Soft X-ray irradiation causes hydrogen around the vacancy-type defect to desorb, resulting in an increase in the size of the vacancy-type defect and an increase in PAL. On the other hand, desorption of hydrogen changes the atoms surrounding the vacancy-type defect to carbon. Since carbon has inner-shell electrons, the fraction of inner-shell electrons that annihilate with positrons is expected to increase, resulting in a decrease in the S-parameter.

### **REFERENCES:**

[1] K. Kanda et al., Sen. Mater., 29 (2017) 817-826.



Fig. 1. The soft X-ray dose-dependence of S-parameter and PAL.

## Hydrogen trapping behavior at vacancy in Fe-Al alloy with electron irradiation

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**INTRODUCTION:** B2 ordered Fe-Al alloy is used as high strength at high temperature material because of its good properties such as specific strength to weight ratio, oxidation resistance. However, it is known that Fe-Al alloy with B2 ordered structure is easy to form vacancy and anti-site atom defects. These defects strongly affect the physical and mechanical properties. So far, we have investigated the nature of defect structures in B2 type Fe-Al alloy. On the other hand, the vacancy type defects in this alloy first principles calculation result indicate that not only one hydrogen atom but also several hydrogen atoms can be trapped in a single vacancy in B2 ordered Fe-Al alloy. However, the interaction between vacancies and hydrogen atoms in this type of Fe-Al alloy is not cleared yet. In this study, we have performed slow positron beam measurements for electron irradiated and hydrogen introduced Fe<sub>50</sub>Al<sub>50</sub> alloys to estimate the hydrogen trapping state at a vacancy.

**EXPERIMENTS:** B2 ordered Fe-Al alloys with stoichiometric composition were prepared by arc melting method in argon gas atmosphere. Sliced samples with the thickness of 0.5 mm were annealed at 1273 K for 20 h and cool down to 973 K slowly and then quenched into water. These specimens were irradiated with 8 MeV electron up to the fluence of  $4 \times 10^{18}$  /cm<sup>2</sup> at KURRI, Kyoto University. Irradiation was carried out at about 330 K with temperature controlling water cooling system. After irradiation, hydrogen was injected by electrochemical method for 8, 16, 80 and 160 hours. These samples were examined by X-ray diffraction and positron annihilation Doppler broadening measurements by using slow positron beam at KURRI facility.

**RESULTS:** Fig. 1 shows the X-ray diffraction peak change of (211) plane after electron irradiation and hydrogen charging. The trends of peak shift due to vacancy generation and hydrogen absorption are opposite. This means that the strain introduced by the formation of vacancies due to electron irradiation was relaxed by the trapping of hydrogen within the vacancies. In addition, as the hydrogen charging time increases, the peak moves to a lower angle compared to that for before irradiation. This

is thought to be due to trapping of multiple hydrogen atoms within the vacancy. Fig. 2 shows the depth profile of positron Doppler S parameter for each hydrogen charging time. As the charging time becomes longer, the S value decreases at a deeper position and the amount of decrease became larger. This result is consistent with the XRD results and suggests that multiple hydrogen atoms may be trapped within one vacancy.





Target Depth, µm

Fig. 1. X-ray diffraction profiles for electron irradiated and hydrogen absorbed Fe-50%Al.

Incident positron energy, keV Fig. 2. The depth profile of S parameter for electron irradiation and hydrogen absorbed samples.

### Structural Changes of Amorphous Germanium Oxide at Shock Compression

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**INTRODUCTION:** The germanium oxide system has been widely studied at high-pressure conditions. This system is an ideal analog of silicate mineral systems, which are major components of terrestrial planets. Silicon and germanium are similar in their valence electron configuration, whereas the larger ionic radius of Ge<sup>4+</sup> relative to Si<sup>4+</sup> facilitates structural changes of the germanium oxides to increase their coordination numbers at lower pressures than the silicates. Understanding the structural changes of germanium oxides are thus important for understanding and predicting the high-pressure behavior of silicates within the planets.

The structure of amorphous GeO<sub>2</sub> consists of interconnected tetrahedra centered by  $Ge^{4+}$  with oxygen coordination number of 4 at ambient pressure. The coordination number gradually increases to 6 in the pressure range up to about 23 GPa under static compression [1]. Under shock compression, both amorphous and crystalline GeO<sub>2</sub> were also expected to have structural changes to increase their coordination number from 4 to 6. However, there have been few structural observations of recovered samples of GeO<sub>2</sub> after shock compression. In this study, therefore, we investigated the structure of shock-compacted amorphous GeO<sub>2</sub>.

**EXPERIMENTS:** The starting sample was prepared by melting GeO<sub>2</sub> crystal powder of 99.999% purity at 1400 °C for 14 hours and at 1450 °C for 6 hours. The recovered sample was crushed and ground with a mortar and pestle made of tungsten carbide, and then pelletized and sealed into a sample container made of SUS304. Shock recovery experiments using these samples and containers were conducted using a single-stage explosive gun at the National Institute for Materials Science [2]. Two experiments were undertaken separately at peak pressures of 17 GPa and 19 GPa. After the sample containers were shock-compressed and recovered, they were cut using a metal blade. Then, one side of the sample was homogenized and analyzed using a powder X-ray diffractometer (Ultima IV). The other side of the sample was analyzed using a focused X-ray diffractometer (SmartLab 9kW) to resolve structures of several points of ~100  $\mu$ m diameters that emerged on the cross-section surface.

**RESULTS AND DISCUSSION:** Fig. 1 shows the powder Amorphous GeO, X-ray diffraction patterns before and after the compression. These samples consisted of 4-coordinated  $\alpha$ -quartz-type and 6-coordinated rutile-type GeO<sub>2</sub> in addition to amorphous matrix. The peak intensity of the  $\alpha$ -quartz type became stronger at 19 GPa; the growth of the  $\alpha$ -quartz type was strongly affected by the peak pressure. The growth of the rutile type was much less sensitive to it. Such a difference suggested that the transformation pathways to the  $\alpha$ -quartz type and the rutile type were independent and coexisted. Whereas the  $\alpha$ -quartz type may be grown in a 4-coordinated amorphous structure in the remaining heat after the pressure release, the rutile type should only be grown in a 6-coordinated amorphous structure during the compression.

### **REFERENCES:**

[1] Y. Kono *et al.*, PNAS, **113** (2015) 3436–3441.
[2] Y. Umeda *et al.* J. Mineral. Petrol. Sci., **119** (2024) 230706.



Fig. 1. X-ray diffraction patterns of samples after shock compression at 17 and 19 GPa and that before the compression.

## Characterization of Precipitates in Cu-Ni-Si alloy using Small-Angle X-ray Scattering

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**INTRODUCTION:** Cu-Ni-Si alloy is one of general precipitation-strengthened Cu alloys and is widely used as electrodes in electronic devices. In this alloy, while nano-sized Ni-Si precipitates are dispersed in Cu matrix and improve the mechanical strength, solute Ni and Si reduce the electric conductivity. Therefore, the precise control of precipitation by aging heat treatment is an important issue. Small-angle X-ray scattering (SAXS) can quantitatively characterize the shape and size of the precipitates in alloys and provide useful knowledge for precipitation mechanism [1-3]. In this study, we performed the SAXS measurements of the Ni-Si precipitates in Cu-Ni-Si alloy with various aging conditions.

**EXPERIMENTS:** The sample was Cu-Ni-Si alloy. The ingot of the alloy was solution-treated and then aged at the temperature between 623 and 773 K. SAXS measurements were performed using the SAXS instrument (NANO-Viewer, Rigaku) with Mo  $K\alpha$  radiation. Scattering patterns were measured using a photon-counting-type two-dimensional detector (PILATUS 100k, Dectris) using a Si converter with the thickness of 1000 µm. The X-ray path from the X-ray entrance slit to the window just before the detector was evacuated to reduce the background scattering from air. The SAXS measurements were performed using two sample-to-detector distances of 0.4 and 1.7 m to cover the *q* range from 0.1 to 9 nm<sup>-1</sup>, where *q* is the magnitude of the scattering vector. The scattering patterns were reduced using our original program written in Igor Pro (WaveMetrics).

**RESULTS:** Fig. 1 shows the SAXS profiles of the solution-treated and aged Cu-Ni-Si alloys. The profile of the solution-treated sample monotonically decreases and has a plateau in the high q region. These are typical characteristics of the solution-treated sample and reflect the scattering of coarse

structures other than precipitates. In contrast, the profile of the aged sample shows a shoulder, which indicates the formation of the nano-sized precipitates with aging. The higher scattering intensity in the solution-treated sample than in the aged sample is due to the larger thickness. We also observed the shift of the shoulder toward lower q with increasing aging time. This means that the size of the precipitates becomes larger with aging. Further analysis is in progress. The size and number density of the precipitates will be compared with the mechanical and electric properties.

- [1] H. Sasaki *et al.*, Mater. Trans., **63** (2022)
   1384-1389.
- [2] T. Saito, J. Alloys Compd., **983** (2024) 173852.
- [3] T. Kumada, J. Nucl. Mater., **528** (2020) 151890.



Fig. 1. SAXS profiles of Cu-Ni-Si alloys. Crosses and circles denote the solution-treated and aged samples, respectively.

## Positron Annihilation Spectroscopy in Neutron-irradiated Fe-Cr Alloys 2

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**INTRODUCTION:** Ferritic stainless and heat resistant steels used as nuclear reactor peripheral materials have high Cr content [1]. In these materials, ductility and toughness remarkably decrease, and hardness and tensile strength increase by aging from 593 to 813 K. This phenomenon is caused by the formation of Fe-rich and Cr-rich phase, and is called 475°C embrittlement [2]. These changes in mechanical properties are an important issue in terms of evaluation of aged deterioration when it was used as a reactor structural material. Positron annihilation spectroscopy (PAS) is very powerful tool to obtain the information of vacancy-type defects (even single vacancies) and precipitates. In Fe-Cr alloys used in this study, positron affinity of Fe is lower than that of Cr [3]. Therefore, we can detect the formation of Fe-rich phase in phase separation of Fe-Cr alloys using PAS. The purpose of this study is to detect the progress of phase separation using PAS, and to obtain the correlation between the hardness and phase separation in Fe-Cr binary alloys irradiated with neutrons at 473K and 573 K.

**EXPERIMENTS:** Fe-x wt.%Cr (x = 0, 9, 15, 30, 45, 50, 70, 85, 91, and 100) binary alloys were used in this study. The weight of high purity Fe (99.99%) and Cr (99.99%) were measured, and samples were melted by the arc melting. For neutron irradiation, samples with diameters of 3 mm and thickness of 0.25 mm were cut using the wire electric discharge machine. Fe-xCr (x = 30, 45, 50, 70, 85, 91, and 100) were annealed at 1273K for 1h, and Fe-xCr (x = 0, 9, 15) were annealed at 1073K for 1h in a vacuum (<  $4 \times 10^{-4}$  Pa), and then water-quenching was performed for the suppression of phase separation. The neutron irradiation was carried out at the Material Controlled Irradiation Facility (SSS) of Kyoto University Reactor (KUR) [4]. The irradiation doses were  $0.44 \times 10^{-3}$ ,  $0.5 \times 10^{-3}$  and  $2.1 \times 10^{-3}$  dpa. The irradiation temperature was 473K ( $0.5 \times 10^{-3}$  dpa) and 573K ( $0.44 \times 10^{-3}$  and  $2.1 \times 10^{-3}$  dpa). Measurements of coincidence Doppler broadening (CDB) were performed at room temperature. The region of S parameter of CDB spectra was defined as  $|p_L| < 4 \times 10^{-3} m_0c$ , and the region of W parameter was defined as  $12 \times 10^{-3} m_0c < |p_L| < 28 \times 10^{-3} m_0c$  ( $m_0$ : rest mass of electrons, *c*: velocity of light).

**RESULTS:** CDB ratio curves in the region of W-parameter heightened after irradiation in Fe-*x*Cr (x = 70, 85, and 91) irradiated at 473 K and in all Fe-Cr alloys irradiated at 573 K. This result indicates that Fe-rich phase grows, and the amount of positrons, which are annihilated with core electrons of Fe atoms, increases. Form PAL measurements, vacancy clusters were not detected in Fe-*x*Cr (x = 70, 85, and 91). These results indicate that almost all positrons are annihilated in Fe-rich phase, and vacancy clusters are not formed in Fe-rich phase. In Fe-*x*Cr (x = 9, 15, 30, 45 and 50) irradiated at 473 K, the decrease in the W-parameter region is caused by the increase in the S-parameter region. After progress of phase separation by neutron irradiation, we cannot obtain the information of defects in Cr-rich phase using positron annihilation spectroscopy.

### **REFERENCES:**

[1] R.L. Klueh et al., J. Nucl. Mater., **191** (1992) 116.

- [2] R.O. Williams, Trans. AIME, **212** (1958) 497.
- [3] M.J. Puska et al., J. Phy. Condes. Matter, 1 (1989) 6086.
- [4] T. Yoshiie et al., Nucl. Instr. Meth. Phys. Res. A, 498 (2003) 522.

## Radiation Properties of Wide-bandgap Semiconductor Sensors for High-Energy Physics Experiments

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**INTRODUCTION:** Wide-bandgap semiconductor sensors have been considered as a potential alternative to Si for the manufacture of dosimeters, spectrometers, and charge particle detectors in high energy physics experiments, by virtue of its operation capability in strong radiation and/or high-temperature environments. To take advantage of such properties for future radiation detectors with a comparable size of silicon, we investigated two different semiconductor materials, SiC and BGaN this year. This study focuses on the influence of the SiC's bulk defects on the radiation sensor characteristics under the reverse-bias condition and neutron-detectability with BGaN sensors, by irradiating neutrons at Institute for Integrated Radiation and Nuclear Science, Kyoto University.

**EXPERIMENTS:** The reverse blocking characteristics and leakage current characteristic are primary concerns of the radiation effects on SiC. On the other hand, BGaN, a mixed semiconductor of GaN and BN, is expected to be a novel neutron detection material because it contains boron (B), a neutron-capturing element, as a semiconductor constituent. Therefore, we conducted different measurements with each sensor. As for the SiC sensor, we irradiated fast neutrons to SiC pn-diodes under the bias condition of 1 kV [1]. Fig. 1 shows the photograph of the measurement setup. We installed two SiC sensors at the front of the rail-instrument. After the 1MW operation, we carried out measurements of the leakage current and compared with those of the pre-irradiation samples. As for the BGaN, the sensor signals have been taken during the irradiation by external DAQ system and signal amplitudes and rise times were evaluated in off-line. The irradiation tests were both conducted at KUR.

**RESULTS:** The typical I-V characteristics of SiC sensors before and after irradiation showed similar characteristics, and we did not observe any degradation in leakage currents. The reverse blocking property was also retained up to 1 kV, which is required for full depletion of the SiC devices. As for the BGaN sensors, neutron-detectability was confirmed with sufficient event rates. Figure 2 is the time vs. neutron's event rate. For irradiations of less than 300 minutes, the counting rate decreased with irradiation time. The counting rate recovered and stabilized after more than 300 minutes of irradiation. This result suggests that neutron irradiation formed damage that affected device properties, but that further irradiation may have recovered the damage. Detailed evaluation of reproducibility and crystallinity dependence is needed to elucidate the mechanism in the future.



Fig. 1. Photograph of the measurement setup.



Fig. 2. Irradiation time dependence of neutron detection counting rate with BGaN detector.

#### **REFERENCES:**

[1] T. Kishishita et al., IEEE Trans. Nucl. Sci., 68 (2020) 2787-2793.

[2] K. K. Lee et al., Nucl. Instrum. Methods Phys. Res. B, 210 (2003) 489-494.

## Effect of gamma radiation on ultra-micro structure of hardwood cell-wall

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**INTRODUCTION:** Space-wood project (LignoStella project) is performed by Kyoto University collaborating with Sumitomo forestry Co Ltd in order to challenge to use wood-material in out-er space. Firstly, possibility to use wood material in outer space has to be confirmed because a small wooden artificial satellite is going to launch until 2024. Space exposure test of wood specimen started on ExBAS at ISS in March 4, 2022. The wood specimen was exposed for about 1 year and came back to Earth in March, 2023. They were affected by atomic oxygen and cosmic ray. In this study, effect of gamma-ray irradiation on cell-wall of wood material was studied on the ground in parallel with the exposure tests in outer space.

**EXPERIMENTS:** The honoki specimens (*Magnolia obovate*) were conditioned in 20°C and 60%RH (dry wood) or 20°C and 97%RH (wet wood) for a few weeks before the irradiation test. To prevent moisture content changes during irradiation, they were wrapped in polyethylene film. The specimens were then irradiated for 24 hours, with three different doses applied: 100 kGy, 14 kGy, and 1 kGy. The fracture properties of the irradiated specimens were measured using a single-edge notched bend (SENB) test conducted on a material testing machine (AG-I/100kN, Shimadzu, Kyoto, Japan). This test measured the load and displacement at the center point.

**RESULTS:** The relationship between fracture energy and fracture toughness values and absorbed dose is shown in Fig. 1. Note that 0 kGy is expressed as 0.01 kGy. Fracture energy in fracture mechanics tests is related to the formation of the fracture progression zone (FPZ), which may reflect the viscosity of the wood. Fig. 1 shows that the fracture energy decreased by 70% at 100 kGy. This suggests that a change in plastic deformation may have occurred at 100 kGy. Murata et al. [1] reported that pyrolysis of wood affects fracture energy. Thermal treatment affects fracture energy of

wood, as the fracture energy of spruce wood after heat treatment decreased linearly from heat treatment above 150°C. The reduction in fracture energy observed in this study aligns with their findings.

Fig. 1 shows that the fracture toughness values exhibited a decreasing trend at 1 kGy and decreased to approximately 80% compared to the control at 100 kGy. Murata et al. [1] reported that the maximum load on heat-treated spruce wood began to decrease with heat treatment above 130°C and remained unchanged beyond 150°C. The fracture toughness values obtained in this study align with the findings of previous research.



Fig. 1. The relationship between fracture energy and fracture toughness values and absorbed dose.

### **REFERENCES:**

[1] T. Aoki *et al.*, bulletin of the Wood Research Institute Kyoto Univ., **62** (1977) 19-28.
[2] K. Murata *et al.*, Materials., **6** (2013) 4186-4197.

## The effect of ion irradiation on $YBa_2Cu_3O_y$ and $FeSe_{0.5}Te_{0.5}$ superconducting films using a slow positron beam

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**INTRODUCTION:** Cuprate and iron-based superconducting films are being developed for use in high magnetic field applications such as generators for wind turbine, magnetically-levitated trains and electric aircraft. For these applications, increasing critical current, which is a maximum value of zero-resistivity current, property in magnetic fields is very important. Critical current properties in magnetic fields are improved by introduction of lattice defects with nano-meter size using ion-irradiation techniques. Positrons are useful for characterizing irradiation-induced defects. Last year, we have probed two kinds of pristine superconducting films, cuprate superconductor YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> (YBCO) and iron-based superconductor FeSe<sub>0.5</sub>Te<sub>0.5</sub> (FST), using a slow positron beam. In this study, we evaluated the irradiation defect in these films using the positron annihilation measurement.

**EXPERIMENTS:** The YBCO(~500 nm) films and the FST(~100 nm) films were irradiated with 10 MeV Au<sup>4+</sup> ( $9.5 \times 10^{12}$  cm<sup>-2</sup>) and 2 MeV Au<sup>2+</sup> ( $4.6 \times 10^{12}$  cm<sup>-2</sup>) ions, respectively. These films were probed by the KUR slow positron beam and the Doppler broadening of annihilation radiation (DBAR) spectra were acquired with incident positron energies  $E_+ = 9$  keV. The sharpness of the DBAR spectra is evaluated by a value called the *S*-parameter, which becomes generally lower when positrons annihilate in a perfect lattice, and higher when positrons are trapped into vacancies [1].

**RESULTS:** Figure 1 shows the *S*-parameters as functions of the incident positron energy for (a) YBCO and (b) FST thin films before and after irradiation. The *S*-parameters for both irradiated samples did not show a clear change, compared with corresponding those for the unirradiated samples. This could be indicative of the presence of vacancy-type defects in high concentration (vacancy concentration:  $C > 10^{19}$  /cm<sup>3</sup>) in both unirradiated samples, so that most of the positrons are trapped at those vacancy-type defects.

We will identify the type of vacancy in these unirradiated films by comparing the momentum distribution of annihilated electrons with the result of the first-principles calculation.

### **REFERENCES:**

[1] R. W. Siegel, Ann. Rev Mater. Sci., **10** (1980) 393–425.



Fig. 1 *S*-parameters as functions of the incident positron energy for (a) 10 MeV Au<sup>4+</sup> irradiated YBCO films and (b) 2 MeV Au<sup>2+</sup> irradiated FST films, respectively.

### Measurements of hyperfine fields in Cd<sub>x</sub>Fe<sub>3-x</sub>O<sub>4</sub>

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**INTRODUCTION:** Spinel ferrites have a characteristic structure comprised of two different sublattices with cubic closest packed oxygen atoms with a crystallographic formula of  $(M^{2+}_{1-s}Fe^{3+}_{s})_A[M^{2+}_{s}Fe^{3+}_{2-s}]_BO^{2-}_{4}$ . Because the main constituent cations are Fe ions, their magnetic property has attracted attention from the view point of application to functional materials. Among a number of candidates of the introduced cations M, we directed our interest to Cd, and investigated Cd-content dependence of hyperfine fields in Cd<sub>x</sub>Fe<sub>3-x</sub>O<sub>4</sub>.

**EXPERIMENTS:** Powder  $Cd_xFe_{3-x}O_4$  of each x was individually synthesized by a conventional solid-state reaction, and characterized the synthesized samples by powder X-ray diffraction patterns [1]. Time-differential perturbed angular correlation (TDPAC) spectroscopy with the <sup>111</sup>Cd( $\leftarrow$ <sup>111</sup>In) probe was applied to the samples to measure the hyperfine fields at the Cd nucleus in the A site. (In our previous work, we found Cd ions specifically occupy the A site by means of the spectrosco-

py with the <sup>111</sup>Cd( $\leftarrow$ <sup>111m</sup>Cd) probe generated in KUR [2].) The room-temperature TDPAC spectra ob-**RESULTS:** tained for the samples of x = 0.1, 0.25, and 0.46 are shown in Fig. 1. The damping structure becomes marked as x increases, indicating the effect of Cd doping on the distribution of the hyperfine field at the probe. We also investigated the temperature dependences of the Larmor frequency of the precession of the <sup>111</sup>Cd( $\leftarrow$ <sup>111</sup>In) probe,  $\omega_L$ , induced by the magnetic hyperfine fields supertransferred to the probes. We observed diminishing trend in the frequencies with increasing temperature, showing activated thermal fluctuation of ordered spins in surrounding Fe ions at higher tempera-The temperature dependences of  $\omega_L$  were able to be ture. reproduced well by the following power law:

$$\forall_{L}(T) = \forall_{L}(0) \overset{\forall}{\underset{\#}{\otimes}} ! \frac{T}{T_{c}} \overset{\exists}{\underset{\%}{\otimes}}.$$
 (1)

Here,  $T_C$  is the Curie temperature. The fits for  $Cd_xFe_{3-x}O_4$  of x = 0.1, 0.25, and 0.46 gave the  $T_C$  values of 806(1) K, 733(3) K, and 658(1) K, respectively. This observation demonstrates that  $T_C$  decreases as the Cd-content x increases, suggesting that the effect of thermal fluctuation of Fe spins is enhanced with increasing Cd content.

### **REFERENCES:**

[1] W. Sato *et al.*, Appl. Radiat. Isotopes, in press.
[2] W. Sato *et al.*, J. Appl. Phys, **120** (2016) 145104 (1-7).



Fig. 1. TDPAC spectra of  $Cd_xFe_{3-x}O_4$ (x = 0.1, 0.25, and 0.46) obtained at room temperature.

## Slow Positron Beam Analysis of Polymer Composite Materials

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**INTRODUCTION:** Membrane treatment technology has the characteristics of low cost, low energy consumption and high efficiency [1]. Polyamide/Polysulfone membrane is one of the popular membranes which have been widely applied in water treatment due to its high salt rejection and excellent water flux. And this membrane has also been applied to treat the low-dose rate radioactive waste water. Considering the irradiation could accelerate the polymer degradation, the radioactive waste water may lead to the faster deterioration of membrane performance. In order to achieve better performance of the separation membrane, it is necessary to study the irradiation effect on the microstructure of the separation membrane. In this study, the polyamide/polysulfone membrane irradiated by an electron beam has been investigated via positron annihilation techniques coupled with a positron beam.

**EXPERIMENTS:** The membranes were prepared via an interfacial polymerization method in the laboratory. And the membranes were irradiated by an electron beam at different doses. Positron measurements were performed with the positron beam system at the Institute for Integrated Radiation and Nuclear Science, Kyoto University. The positron annihilation Doppler broadening was expressed by the *S* parameter, which is defined as the ratio of annihilation photon counts in a predetermined window centered at 511 keV to the total counts in the annihilation peak.

**RESULTS:** As shown in Fig. 1, the layer structure of the four membranes can be distinguished from the *S*-*E* curves. Here *E* denotes the incident positron energy; the higher the *E*, the deeper the positrons are injected. It can be seen from Fig. 1 that, the *S*-*E* curves can be divided into two stages, a surface polyamide layer ( $0 \sim 3 \text{ keV}$ ) and a polysulfone underlayer ( $3 \sim 20 \text{ keV}$ ). The *S*-*E* shape is similar to the positron results of polyamide/polysulfone membranes in the previous study [2]. Besides, one can find that the *S* parameters decrease when the membrane irradiated at higher dose. The decrease of the *S* parameters might be attributed to the formation of polar groups formed during the irradiation process.

- [1] J. Yin and B. Deng, J. Membrane Sci., 479 (2015) 256-275.
- [2] Z. Chen et. al., J. Phys. Chem. C, 115 (2011) 18055-18060.





# Effects of lead addition on tritium recovery for Li<sub>2</sub>TiO<sub>3</sub>-Li<sub>4</sub>SiO<sub>4</sub> mixed breeder material

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### **INTRODUCTION**

Tritium will be produced by  $(n, \alpha)$  reaction with lithium (Li) in the fusion reactor because the amount of tritium in nature is small. Tritium breeder needs good chemical stability and easy to recover tritium. Lithium silicate (Li<sub>4</sub>SiO<sub>4</sub>) and lithium titanate (Li<sub>2</sub>TiO<sub>3</sub>) have different characteristics. For example, lithium silicate has a high lithium density, while lithium titanate is good chemical stability and has a good crushing load. Advanced tritium breeder materials should have both of higher tritium production rate and recovery efficiency. LTO-LSO mixed ceramic materials are expected to have both advantages. Lead (Pb) is a neutron multiplier that contributes to the improvement of the tritium breeding ratio and has a lower affinity with tritium. The addition of Pb into Li ceramics is expected to have a potential advantage for efficient tritium recovery.

### **EXPERIMENTS**

LTO-LSO-5wt.%Pb, a powder sample of lithium titanate and lithium silicate mixture with 5wt.%Pb addition, was prepared at Wuhan University of Technology. Those samples were irradiated by neutron at Kyoto University Research Reactor (KUR) with the neutron fluence of  $8.0 \times 10^{15}$  n cm<sup>-2</sup> and  $8.0 \times 10^{16}$  n cm<sup>-2</sup>. After the neutron irradiation, tritium release behavior was evaluated by Tritium-thermal desorption spectroscopy (T-TDS) system by heating the samples separately from R.T. to 1113 K with the heating rates of 10, 20, and 30 K min<sup>-1</sup>. Liquid scintillation counter (LSC) was used to measure the total T amount trapped by the water bubbler at Shizuoka University.

### **RESULTS AND DISCUSSION**

Fig. 1 shows the T-TDS spectra for LTO-LSO-5wt.%Pb with two different

neutron fluences. The amount of tritium released was increased as the fluence was increased. The sample with the low neutron fluence of  $8.0 \times 10^{15} \text{ n} \cdot \text{cm}^{-2}$ showed a large desorption stage located at a lower temperature side of 400 K, whose activation energy was 0.15-0.33 eV, ascribing to the desorption of tritium adsorbed on the surface. As the fluence increases, the desorption of tritium from the bulk was the major tritium desorption stage. Fig. 2 shows the T-TDS spectra for mixed sample without Pb with neutron fluence of  $8.0 \times 10^{16}$  $n \cdot cm^{-2}$  at 20 K min<sup>-1</sup>. Compared to the mixed sample without Pb in the pebble sample and the mixed sample including Pb in the powder sample, the major desorption stage was shifted to the lower temperature side. This is due to more efficient tritium recovery with the addition of Pb. In this experiment, it was found that the amount of irradiation defects caused by neutron irradiation decreased with the addition of Pb. It can be said that the number of trap sites for tritium desorption in the bulk decreased. Therefore, it is considered that the addition of Pb reduces the number of trapping and de-trapping processes, and thus tritium recovery can be achieved at low temperatures. The TDS spectrum did not change significantly, suggesting that the process of tritium desorption should be the same.

### REFERENCES

[11] S. Hirata et al., J. Nucl. Mater. (2022) 1538383.



Fig. 1 TDS spectra of LTO-LSO-5wt%.Pb with the neutron fluence of  $8.0 \times 10^{15}$  and  $8.0 \times 10^{16}$  n • cm<sup>-2</sup>.



Fig. 2 TDS spectra of 2LSO-LTO, LSO-LTO and LSO-2LTO with the neutron fluence of  $8.0\times10^{16}$  n  $\cdot$  cm^{-2} at 20 K min^{-1} [1].

## Effect of rhenium doping on vacancy formation in electron-irradiated tungsten studied with positron annihilation spectroscopy

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**INTRODUCTION:** Tungsten (W) is promising as a plasma-facing material for fusion reactors, and material development is ongoing both domestically and internationally. Although W has the lowest hydrogen solubility limit among metal elements, the retention and accumulation of hydrogen isotopes can become a problem under irradiation. However, it has been reported that adding rhenium (Re) significantly reduces the accumulation of hydrogen isotopes [1]. This is thought to be due to the suppression of the formation of irradiation-induced defects, which serve as hydrogen trapping sites, by the addition of Re. Indeed, we reported that the formation of vacancy-type defects is significantly suppressed in W-5Re compared to pure W using neutron-irradiated or electron-irradiated W alloys. This study aims to further investigate the effect of Re addition on the formation of vacancies by introducing only simple Frenkel pairs through electron irradiation in W-Re dilute alloys and examining vacancy-type defects using positron annihilation spectroscopy.

**EXPERIMENTS:** The test materials are pure W and W-xRe alloys (x = 0.1, 0.3, 1.0, 5.0 at.%). After annealing at 1800°C for one hour (1600°C for one hour for x = 5.0), electron irradiation (8 MeV, <100°C, approximately  $3.5 \times 10^{23}$  e<sup>-</sup>/m<sup>2</sup>) was carried out at the Kyoto University Linac. After irradiation, vacancy-type defects were examined using positron lifetime measurements.

**RESULTS:** Figure 1(a) shows the Re composition dependence of the average positron lifetime  $(\tau_{ave})$ . In the unirradiated samples,  $\tau_{ave}$  ranged from 110 to 117 ps. Since these values are close to those of bulk W, it is believed that there were almost no vacancy-type defects serving as positron trapping sites before irradiation. After irradiation,  $\tau_{ave}$  increased for all samples compared to before

irradiation, indicatingpositron trapping in irradiation-induced vacancy-type defects. For all samples, the positron lifetime spectra exhibited little to no long-lived component, suggesting minimal positron trapping in vacancy clusters. Figure 1(b) shows the increase in  $\tau_{ave}$  ( $\Delta \tau_{ave}$ ) due to irradiation. Compared to pure W,  $\Delta \tau_{ave}$  increased with Re composition up to about 1%, indicating an increase in positron trapping in vacancy-type defects. However, at 5% Re composition,  $\Delta \tau_{ave}$  was smaller than in pure W, consistent with the previous report on W-5Re. In that previous study, the mechanism proposed was that Re strongly binds to W interstitial atoms, reducing their mobility and increasing the frequency of interstitial-vacancy recombination, thereby suppressing the formation of vacancy-type defects [2]. However, the results of this study suggest the need to consider other mechanisms. For example, Re has a positive (attractive) binding energy of 0.22 eV with vacancies in W [3], suggesting the possibility that it stabilizes vacancies. **REFERENCES:** 



Fig 1. (a)Average positron lifetime in pure W and W-Re alloys before/after electron irradiation. (b) Increase of the average positron lifetime in pure W and W-Re alloys.

- [1] J. Wang et al., J. Nucl. Mater., 559 (2022) 153449.
- [2] T. Suzudo et al., Sci. Rep., 6 (2016) 36738.

### Dependence of In Concentration on the Local Structure of In Doped in SrTiO<sub>3</sub>

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**INTRODUCTION:** SrTiO<sub>3</sub> is one of the most promising candidates for photocatalytic materials. It is known that trivalent cation doping at the Ti<sup>4+</sup> site is one of the effective methods to improve photocatalytic activity. According to previous research, it is known that doping trivalent cations such as  $In^{3+}$  into Ti<sup>4+</sup> suppresses the formation of Ti<sup>3+</sup> recombination centers and improves the photocatalytic activity of SrTiO<sub>3</sub>. On the other hand, if a secondary phase of indium oxide is formed due to excessive doping, it is thought that the secondary phase becomes a recombination center and the photocatalytic activity decreases. For the precise control of photocatalytic properties of In-doped SrTiO<sub>3</sub>, therefore, it is necessary to examine the local surroundings of  $In^{3+}$  dopant of various In concentrations. For the microscopic study of the local fields at In sites, TDPAC measurements were performed for  $^{111}Cd(\leftarrow^{111}In)$  in undoped and In-doped SrTiO<sub>3</sub>.

**EXPERIMENTS:** SrTiO<sub>3</sub> powders doped with 0.1, and 1 at.% In were synthesized by the conventional solid-state reaction as described in the references[2]. Commercially available <sup>111</sup>In HCl solution was added in droplets onto the disks of undoped SrTiO<sub>3</sub> and each In-doped SrTiO<sub>3</sub> sample. After the disks were dried up by heat, they were heated in air at 1473 K for 24 h for the diffusion of the probe. Each disk was then ground into powder and sealed in a pyrex tube. TDPAC measurements were performed at room temperature by the detection of the 171–245-keV cascade  $\gamma$  rays with the intermediate spin I = 5/2 having a half-life of 85.0 ns

**RESULTS:** Figure 1 shows the TDPAC spectra of the <sup>111</sup>Cd( $\leftarrow$ <sup>111</sup>In) probe embedded in undoped, 0.1 at.%, and 1 at.% In-doped SrTiO<sub>3</sub> measured at room temperatures. From the fitting parameters of the TDPAC spectrum in Fig. 1(a), it is suggested that the <sup>111</sup>Cd( $\leftarrow$ <sup>111</sup>In) probe is uniformly doped into SrTiO<sub>3</sub> and occupies three types of sites. Considering the previous study,  $In^{3+}$  ions occupy the B sites in  $SrTiO_3[1]$ . Therefore, there would be three types of local structures in the vicinity of the  $^{111}Cd(\leftarrow^{111}In)$ probes at the substitutional Ti<sup>4+</sup> site. For Fig.1(b), the parameters of quadrupole frequencies between the two results of Fig.1(a) and (b) were the same within the fitting error range. Therefore, it is suggested that 0.1% of  $In^{3+}$  is uniformly doped into SrTiO<sub>3</sub>. However, in the TDPAC spectrum shown in Fig. 1(c), components derived from the secondary phase of In<sub>2</sub>O<sub>3</sub> were observed, which suggests that  $In^{3+}$  partially forms  $In_2O_3$  in 1 at.% In-doped SrTiO\_3. These results suggest that in the case of In-doped SrTiO<sub>3</sub> synthesized by the conventional solid-state reaction, the solubility limit of  $In^{3+}$  is between 0.1 and 1 at.%.

### **REFERENCES:**

[1] H. Sudrajat *et al.*, Phys. Chem. Chem. Phys., **22** (2020) 19178-19187.

[2] S. Komatsuda et al., Interactions, 245 (2024) 37.



Fig. 1. TDPAC spectra of  $^{111}Cd(\leftarrow^{111}In)$  in undoped and In-doped SrTiO<sub>3</sub> samples.

## Sample Preparation for Study on HPLC Elution Behavior of Heavy Lanthanide Metallofullerene Using Chlorobenzene Eluent

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**INTRODUCTION:** Lanthanide (Ln) metallofullerene (EMF): Ln@C<sub>82</sub> is a clathrate compound encapsulating metal atom in fullerene C<sub>82</sub> and known that two or three electrons are transferred to C<sub>82</sub> cage from the encapsulated Ln atom [1]. In the previous work, we succeeded to obtain the high performance liquid chromatography (HPLC) elution behavior on a pyrenyl stationary phase for Ln@C<sub>82</sub> of La, Ce, Pr, Nd, Gd, Tb, Dy, Ho, and Er with a toluene as an eluent by the thermal neutron activation method [2]. In order to obtain further knowledge about the HPLC elution behavior of Ln@C<sub>82</sub>, we used chlorobenzene (CB), which has a similar structure to toluene, as an eluent in this study. In general, the neutron activation of compounds containing Cl is required some precaution. In particular, since Dy which has a short half-life (<sup>165</sup>Dy:  $T_{1/2} = 2.3$  h) is expected to be affected very much, we investigated the influence of <sup>38</sup>Cl on the measurement prior to this study.

**EXPERIMENTS:** Ln@C<sub>82</sub>s of Dy, Ho, and Er were prepared by previously reported [2]. These purified samples were injected into a Buckyprep column using CB as an eluent. The eluate from the column was fractionated every 2 minutes at room Temperature (RT) and every 30 seconds at 50 °C. These fractionated eluates were evaporated to dryness and re-dissolved to carbon disulfide and then dropped onto paper filters with 12 mm diameters and dried for 2 or 3 days in the evacuated desiccator. After sufficient drying, these samples were measured XRF spectra to confirm the amount of Cl on the filters and sealed into polyethylene bags and irradiated in ethylene vial. These sealed samples were activated by a thermal neutron in the KUR of the Institute for Integrated Radiation and Nuclear Science, Kyoto university. After the irradiation, the  $\gamma$  rays emitted from the samples were measured by a Ge detector.

**RESULTS:** As the results of XRF, characteristic X ray derived from Cl on the filters was not observed with the significant intensity for almost all samples. The averaged count rate of the observed  $\gamma$ -ray from <sup>38</sup>Cl at 1643KeV for each activated sample was 15.6±2.6 cps for all samples and was found that it had almost no effect on the  $\gamma$ -ray measurement of activated Ln@C<sub>82</sub>. Figure 1 shows the HPLC chart of heavy Ln@C<sub>82</sub> (Ln: Dy, Ho, and Er) with CB as eluent at RT (top) and 50 °C (bottom). In both chromatograms, the HPLC elution behavior of Dy, which had a short half-life and was expected to be most affected by <sup>38</sup>Cl, was confirmed with sufficient intensity. Therefore, it was found that it is possible to use as an eluent containing chlorine by sufficient drying.

### **REFERENCES:**

H. Shinohara, Rep. Prog. Phys., 63 (2000) 843-892.
 K. Akiyama *et al.*, KURNS Prog. Rep., 2022 CO4-1.



Fig. 1. HPLC chart of heavy  $Ln@C_{82}$  (Ln: Dy, Ho, and Er) with CB as eluent at RT (top) and 50 °C (bottom).

## Valuation of Surface Damage on Si induced by the Plasma Treatment using the Positron Annihilation Method

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**INTRODUCTION:** We have been studying the effect of the surface damages, which are induced during the plasma treatment on Si substrates, using the positron annihilation method. In the present study, the effect of the plasma species with the different plasma condition of our previous study [1] and the ion-irradiation damage on Si surfaces during the SF<sub>6</sub>-plasma processing were investigated.

**EXPERIMENTS:** The Si substrates with the size of 18 mm x 18 mm were prepared by cleavage of a Si (100) wafer. To study the effect of the plasma species, each Si substrate was exposed to SF<sub>6</sub>, Ar or CHF<sub>3</sub> plasma (gas pressure: 10 Pa, RF power: 40 W, exposed time: 30 min.). To study the effect of the ion irradiations during the plasma processes, SF<sub>6</sub> plasma (30 Pa, 100 W, 30 min.) were exposed to two Si substrates, the one (SF<sub>6</sub>-1) was positioned on the sample stage, which is connected to blocking condenser and negatively charged during the plasma exposure, and the other (SF<sub>6</sub>-2) was positioned on the cover of the chamber, which is connected to ground. After the plasma treatment, the S-parameters obtained from the Doppler broadening for the energy of the  $\gamma$ -ray by the positron-electron annihilation were measured for all samples.

**RESULTS:** Figure 1 shows the S-parameters for samples with different plasma species as a function of positron energy. The result is almost the same tendency with our previous results [1] (30 Pa, 5 W) except for the result of CHF<sub>3</sub> plasma, which was also observed for BF<sup>2+</sup> ion implanted Si [2]. This might be because of the sputtering effect of the C-related material, which is deposited on the surface by the dissociation of CHF<sub>3</sub>, by ions due to larger RF power in this study. From Fig. 2, it is found that the S-parameters of SF<sub>6</sub>-1 was larger than those of SF<sub>6</sub>-2, indicating that the ion irradiation induces the damages, such as vacancies, near the surface region during the plasma treatment.

### **REFERENCES:**

[1] J. Yanagisawa et al., KURNS Progress Report 2022 (Kyoto University) 112 (CO4-21).

[2] A. Uedono et al., Jpn. J. Appl. Phys. 36 (1997) 969.



Fig. 1. S-parameters of Si exposed to plasma using various kind of gas species at gas pressure of 10 Pa and RF power of 40 W for 30 min.



Fig. 2. S-parameters of  $SF_6$  plasma (gas pressure: 30 Pa, RF power: 100 W, 30 min.) exposed Si substrate positioned on the sample stage ( $SF_6$ -1) and the cover of the chamber ( $SF_6$ -2).

## Study on <sup>99m</sup>Tc separation/concentration technolog from <sup>99</sup>Mo by (n, $\gamma$ ) method (2)

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**INTRODUCTION:** The research and development have been carried out to produce <sup>99</sup>Mo by the  $(n, \gamma)$  method in terms of nuclear security. To apply the  $(n, \gamma)$  method to a <sup>99</sup>Mo/<sup>99m</sup>Tc generator, it is necessary to improve the adsorption properties of alumina (Al<sub>2</sub>O<sub>3</sub>) used as a Mo adsorbent. In this work, several types of alumina were synthesized based on two references that mentioned alumina with excellent adsorption properties and their applicability to the generator was evaluated.

**EXPERIMENTS:** MoO<sub>3</sub> pellet pieces were irradiated with Pn-2 and dissolved with 6M-NaOHaq. The solution was adjusted to a Mo concentration of 10 g/L and pH4. Three types of alumina were prepared: the alumina (Al<sub>2</sub>O<sub>3 4:1</sub>, Al<sub>2</sub>O<sub>3 6:1</sub>) calcined at 300 °C with raw material ratios of 4:1 and 6:1 based on reference [1], and the alumina (Al<sub>2</sub>O<sub>3 HMMA</sub>) based on reference [2]. The Mo solution was added to approximately 0.3 - 0.5 g of the alumina and left for 3 hours, then the alumina was washed with water and transferred to a column. Saline was run through the alumina-filled column to remove <sup>99</sup>Tc. For milking, 10 mL of saline was flowed through the columns every 24 hours for 2 days. The radioactivity of the obtained solutions was measured using a  $\gamma$ -ray spectrometer.

**RESULTS:** Figure 1 shows SEM images of each alumina. For  $Al_2O_{3 4:1}$  and  $Al_2O_{3 6:1}$ , an agglomerated surface of fine particles was observed. On the other hand,  $Al_2O_{3 HMMA}$  had uniform pores on the surface. Table 1 shows the Mo adsorption capacities and <sup>99m</sup>Tc elution rates, <sup>99</sup>Mo/<sup>99m</sup>Tc ratios for each alumina. In the literature, the Mo adsorption capacity of  $Al_2O_{3 HMMA}$  was reported to ex-

ceed 200 mg-Mo/g, but the highest Mo adsorption capacity in the test results was 98.3 mg-Mo/g for Al<sub>2</sub>O<sub>3 6:1</sub>. The elution rates of <sup>99m</sup>Tc were almost the same for all alumina. <sup>99</sup>Mo/<sup>99m</sup>Tc, which is an indicator of <sup>99</sup>Mo desorption rate from the column, deviated from the standard value ( $\leq 0.015\%$ ). The <sup>99</sup>Mo desorption rate from Al<sub>2</sub>O<sub>3 HMMA</sub> was particularly high. Therefore, under this experiment conditions, the surface condition of the alumina may have little effect on Mo adsorption and large effect on the ability to retain Mo. In the future, we will evaluate its applicability to the generator under more practical conditions, where Mo is adsorbed with the column packed with alumina.



Fig. 1. The SEM images of each alumina. (a)  $Al_2O_{34:1}$ , (b)  $Al_2O_{36:1}$ , (c)  $Al_2O_{3HMMA}$ 

Table 1. <sup>99</sup>Mo adsorption capacities of the Alumina, <sup>99m</sup>Tc elution rates and <sup>99</sup>Mo/<sup>99m</sup>Tc rates at milking

				<u> </u>
Sample	<sup>99</sup> Mo adsorption capacity (mg-Mo/g)	Day	<sup>99m</sup> Tc elution rate (%)	<sup>99</sup> Mo/ <sup>99m</sup> Tc (%)
$Al_2O_3_{4:1}$	90.7	1	76.8	1.545
		2	69.0	1.132
Al <sub>2</sub> O <sub>3 6:1</sub>	98.3	1	79.4	1.889
		2	69.8	1.361
Al <sub>2</sub> O <sub>3 HMMA</sub>	91.6	1	77.7	8.008
		2	72.1	2.961

### **REFERENCES:**

X. Hu *et al.*, Mater. Res. Bull., **47** (2012) 4271–4277.
 J. Wang *et al.*, Appl. Radiat. Isot., **178** (2021) 109986.

## Observing the effect of deuterium charge in the modeled metal bilayers of hydrogen embrittlement-resistant alloys

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### **INTRODUCTION:**

As the technology for lightweighting structural metal materials and hydrogen infrastructure are being developed for the purpose of global environmental protection, measures to prevent hydrogen embrittlement of high-strength structural materials are an urgent issue. Elucidation of the mechanism of hydrogen embrittlement is essential for the development of high-performance steels with high strength and resistance to hydrogen embrittlement.

One kind of hydrogen embrittlement-resistant alloys has a system in which nano-precipitates (such as TiN and TiC) are dispersed in steel matrix [1,2]. In this study, we aim to elucidate the behavior of hydrogen trapping by using a metallic multilayer film that models the interface between the nanoprecipitates and the matrix.

### **EXPERIMENTS:**

The metal-multilayer sample was deposited by using ion beam sputtering at KURNS [3]. The sample consisted of 20 repetitions of the bilayer of Fe (0.5 nm) and Ti (0.5 nm) on glass substrate. Because the multilayers of Fe and TiN did not show any significant changes by hydrogen/deuterium charge in the preceding study, we modified the material of TiN layer to Ti because one of hydrogen storage materials is FeTi(H). The Ti-layers were sputtered under hydrogen atmosphere. The sample area was  $40x50 \text{ mm}^2$ .

Deuterium(D) was charged to the multilayers by cathodic electrolysis at a current density of 2  $mA/cm^2$  for 30 minutes using a deuterium aqueous solution of 3mass% NaCl + 0.01mol/l KSCN.

Prior to the neutron reflectivity measurement (NR) at the MINE beamline, the thicknesses of multilayers were determined by X-ray reflectivity (XR) measurement. The results showed that the total thickness of the pair of Fe and Ti(H) changed from 1.18 nm to 1.23 nm after the D charge, which corresponds to 4% increase of total thickness.

Finally, we conducted NR measurement at C3-1-2 (MINE), JRR-3 to test its sensitivity to the change by D charge by observing the critical angle that define the Scattering Length Density (SLDs) of layers.

### **RESULTS:**

NR measurement on the multilayer sample showed the significant increase in the critical angle between before and after the D charge. Those results from XR and NR confirmed that significant amount of D was charged in [Fe/Ti(H)]x20 multilayers and XR and NR are suitable tools. The next step is to measure the difference of SLD between D and hydrogen (H) by NR. We will continue further study for optimizing the ideal model multilayers to identify the location and status of charged D or H.

- [1] Takenori Nakayama et al., Materia, **41(3)** (2002) 230.
- [2] Japanese Patent No.2932943, etc.
- [3] M. Hino, et al., Nucl. Instr. and Meth., 797(2015) 265.

## **Complex Structure of Ions Coordinated with Hydrophilic Polymer 24.** - Obstruction of Iodine Doping by Solute.

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**INTRODUCTION:** We have been investigating dynamical and interacted structures between iodine and polymers. Present "iodine" is "polyiodide ions" ( $I_n^{m-}$ , *m*, *n*: integer, n > 1: "Poly-Iod.(s)" mentioned below), which are defined as charged molecules constructed with more than one iodine And, "Poly-Iod.(s)", mono iodide ion ( $I^{-}$ ) and iodine ( $I_{2}$ ) can actively interact with each othatom. er as molecular (re)structuring between (or among) them. While such (re)structuring can be observed in coordination within organic crystals, it is more emphasized through interaction with mac-We had observed diffusion, coordination and (re)structuring of romolecular matrices.[1] "Poly-Iod.(s)" in some hydrophilic polymers, such as polyamide-6 (PA6); there are observed both rapid diffusion of "Poly-Iod.(s)" into polymers and restructuring as coordinated materials while diameter of iodine atom (or "Poly-Iod.(s)") is relatively large, c.a. 0.4nm.[2,3] Through past researches of characteristic behaviors in coordination of iodine in polymers, we are clarifying unexpected discovery behind simple operation and generality in popular experiments, such as "iodine-starch reaction".

**EXPERIMENTS:** Keeping proportion between I<sup>-</sup> (potassium iodide, KI) and I<sub>2</sub> as 3:1, solutes of "Poly-Iod.(s)" were prepared as "0.2N": i.e., 0.1 mol/L for I<sub>2</sub> and 0.3mol/L for KI. On the other hand, composition of solutes for 3:1 "iodine (1st) doping" were prepared with H<sub>2</sub>O and ethanol (EtOH) as H<sub>2</sub>O : EtOH = 4:0 ("Et:0") / 3:1 ("Et:25") / 1:1 ("Et:50") / 1:3 ("Et:75"). As "iodine (1st) doping" process, dried PA6 films ("Rayfan" in 0.1mm thickness, TORAY) were immersed in the "Poly-Iod.(s)" solutes for more than 3 weeks at R.T. After "iodine (1st) doping" as termination, iodine-doped samples were rinsed with mixture solutes of H<sub>2</sub>O and EtOH without iodine as similar proportion ("Et:0" / "Et:25" / "Et:50" / "Et:75") as doping solutes,

**RESULTS AND DISCUSSION:** As consequence, mass increasing with iodine doping ( $\Delta m$ ) are lowered with proportion of ethanol of solutes: i.e.,  $\Delta m(\text{Et:0}) > \Delta m(\text{Et:25}) > \Delta m(\text{Et:50}) > \Delta m(\text{Et:75})$ ; composition of ethanol prevents "iodine (1st) doping". And, past research with modifying concentration of KI also shows that, even with constant [I<sub>2</sub>], increasing [I<sup>-</sup>] prevents "iodine (1st) doping".[4] If only solved concentration of iodide ion (I<sup>-</sup>) and iodine (I<sub>2</sub>) could control diffusion of "Poly-Iod.(s)" and structures after "iodine (1st) doping", neither composition of solutes (ethanol ratio) nor additional [I<sup>-</sup>] would influence results; effects by "Et:xx" and [I<sup>-</sup>] actually indicate that complicated mechanism behaves as interaction between "Poly-Iod(s)" or with polymers.

Strictly speaking on definition, "Poly-Iod(s)" should include neither (mono) iodide ion,  $I^-$ , which has been charged but is constructed by one iodine atom, nor simple iodine,  $I_2$ , which does not have charge. Nevertheless, "Poly-Iod(s)" interact with both  $I^-$  and  $I_2$  actively. Furthermore, there can be dynamical structures constructed and modified among them; anisotropic structuring among "Poly-Iod(s)" is achieved through characteristic coordination between iodine atoms. Such interaction or structuring is called "iodine bonds" and is attributed to anisotropic linking between "Poly-Iod(s)" or " $\sigma$ -hole" which is most conspicuous positive charge deviation in arrays comparing with other halogen atoms. "Modified orientation" reported previously should also be explained by such dynamism.

- [1] T.J. Marks & D.W. Kalina, "Extended Linear Chain Compounds" vol.1 ch.6., ed. J.S. Miller (Plenum Press, 1982).
- [2] A. Kawaguchi, Polymer, 35 (1994) 3797-3798.
- [3] A. Kawaguchi et.al., SPring-8 User Exp. Rep. 5 (2000) 354-354.
- [4] A. Kawaguchi, Polymer, **37** (1996) 4877-4880.

## Gamma-ray induced light emission from imprity doped GaP single crystal

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**INTRODUCTION:** Gallium phosphide (GaP) is usually employed in the manufacture of low-cost red, orange, and green light-emitting diodes (LEDs). We recently succeeded in detection of the gamma-ray induced photo emission from GaN [1] and ZnO [2] single crystal wafers. It is interesting topic that the light-emission by the excitation of gamma-ray at room temperature in GaP as similar to GaN and ZnO. This is because it can be used as an application as a gamma-ray detector. In the present study, we investigate the emission properties in the gamma-ray excitation to two GaP crystal samples, which is impurity doped and undoped crystal to reveal the role of the impurities for the light emission.

**EXPERIMENTS:** GaP single crystal wafers with a size of several cm square and a thickness of several 100  $\mu$ m meters were used for the present study. The crystals were irradiated at room temperature with gam-ma-rays of 1.17 and 1.33 MeV from a cobalt-60 source of Institute for Integrated Radiation and Nuclear Science, Kyoto University. The gamma-ray irradiation induced photo emission measurements were performed by using a charge coupled device (CCD) equipped spectrometer (QE Pro, Ocean Insight Co. Ltd.).

**RESULTS:** Fig. 1 shows the gamma-ray induced photo emissions for un-doped and impurity doped GaP single crystal samples. These emissions were measured at room temperature with an absorbed dose rate of about 0.8 kGy/h us-ing GaP single crystal wafer with 200  $\mu$ m in thickness. The gamma-ray induced photo emission from impurity doped GaP was observed from 750 nm to 1000 nm, with a peak at around 850 nm and a half width of 250 nm. The greater width of the spectra with the greater width observed in GaP has been attributed to donor-acceptor pair transitions. These spectra are usually broad because of strong phonon-assisted transitions arising from the tight binding of the O donor. The donor – acceptor pair transition energy hv is rep-resented by,

$$hv = E_g - (E_D + E_A) + e^2/4\pi\varepsilon_0\varepsilon_r r,$$

where  $E_g$  is the energy band gap (2.26 eV at 300 K [3]),  $E_D$ and  $E_A$  the isolated donor and acceptor binding energies,  $\varepsilon_0$ the static dielectric constant (11.1), r the distance between the isolated donor and acceptor. Using 0.893 eV as  $E_D$  for O in P site [4], 0.204 eV as EA for Si in P site [5], and r =3.853 Å as the nearest neighbor distance of P site, hv is calculated to be 1.4998 eV (826 nm). This calculated wavelength is close to the observed peak. Therefore, the gamma-ray induced photo emission observed in GaP would be attributed to the O donor – Si acceptor pair transition. On the other hand, the gamma-ray induced photo emission was not observed in un-doped GaP wafers, suggesting that the existence of impurity is important for the photo emission.

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Fig. 1. Gamma-ray-induced photoemission spectra of impurity doped, and un-doped GaP single crystal wafers.

- [1] T. Nakamura et al., Appl. Phys. Lett., 118 (2021) 032106.
- [2] T. Nakamura et al., Solid State Commun., **336** (2021) 114413.
- [3] M. R. Lorenz et al., Phys. Rev., 171 (1968) 876.
- [4] P. J. Dean et al., Phys. Rev., 168 (1968) 812.
- [5] P. J. Dean et al., J. Appl. Phys., **39** (1968) 5631.

### A study on destruction of cesium aluminosilicate compounds by gamma irradiation (5)

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**INTRODUCTION:** Pollucite which is one of cesium aluminosilicate compounds have attracted attention as a final storage material of <sup>134</sup>Cs and <sup>137</sup>Cs. Pollucite is easily synthesized by hydro-thermal method in low temperature below 300°C [1]. Pollucite has various properties that favor the immobilization of Cs ions. However, the damage to the aluminosilicate framework by radiation decay is concerned because it contains <sup>134</sup>Cs and <sup>137</sup>Cs. It has been reported that the effect of  $\beta$ -ray emission and nuclide conversion by  $\beta$ -decay of <sup>137</sup>Cs on aluminosilicate framework is minor [2, 3]. On the other hand, there are few reports of effects by gamma rays on pollucite framework. Therefore, we examined the effect of gamma radiation on the aluminosilicate framework of Pollucite. In this report, we have studied on cesium leakage from pollucite with varying irradiation doses.

**EXPERIMENTS:** Sodium aluminate, sodium metasilicate and cesium chloride were dissolved in sodium hydroxide solution. The solution was placed in a Teflon inner cylinder pressure container. Pollucite was synthesized by hydrothermal method, holding the container at 180°C. The resulting precipitate was washed by distilled water or/and hot water. Thereafter, each solid was collected by filtration and dried at 110°C for 12 hours or more. Hydrothermal syntheses were also performed by replacing the sodium salts of the above reagents with potassium salts.

The powder samples were characterized by XRD, and gamma-irradiated with varying doses by <sup>60</sup>Co source. The leaching test by PCT-A method [4] was carried out to evaluate the change of Cs

retention performance by framework damage. Concentration of cesium in solution leached was estimated by atomic absorption spectrophotometry.

**RESULTS:** Figure 1 shows the results of leaching tests performed with various conditions. As a result of the Fig.1, we found that the leakage of cesium from pollucite depends largely on the cleaning procedure but not on the irradiation dose.

### **REFERENCES:**

[1] Y. Yokomori *et al.*, *Sci. Rep.*, **4** (2014) 4195.



Fig. 1. Results for leaching tests of cesium from pollucite with varying doses by  $^{60}$ Co source.

[2] J. Fortner et al., Argonne National Laboratory, Argonne, Illinois 60439 (2001).

Leaching rate (%)

[3] N. J. Hess et al., J. Nucl. Mater., 281 (2000) 22-33.

[4] ASTM C 1285-02 (2008).

## Study on the porosity of DLC coatings with different microstructures by positron annihilation spectroscopy

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**INTRODUCTION:** The increasing need to enhance the operational efficiency of mechanical systems has brought an increased focus on low-friction coatings. In response, diamond-like carbon coatings (DLCs) have garnered significant attention due to their exceptional mechanical and tribological properties [1]. Specifically, the microstructure of DLC coatings can be modified to meet various industrial demands; their possible structures can extend to graphite-like carbon (GLC) and polymer-like carbon (PLC) by altering deposition conditions and fabrication methods [2]. These structural characteristics are determined by factors such as hydrogen content and the ratio of carbonaceous bonds ( $sp^2$  and  $sp^3$ ). However, studies have indicated that defects also play a role in influencing the characteristics of coatings [3]. Positron annihilation spectroscopy (PAS) is a useful method for elucidating the defect behavior of materials. This study aims to investigate the porpsity of the DLC coatings with different microstructures using PAS.

**EXPERIMENTS:** The hydrogenated (H-) and fluorinated (F-) DLC coatings were fabricated by a plasma-based ion implantation (PBII) technique. The coatings were prepared onto Si (100) wafers. Different negative bias voltages, ranging from -1 to -10 kV, were directly applied to a target as a deposition parameter to vary their microstructure. The deposition period was adjusted to have a thickness of approximately 1  $\mu$ m. The behavior of defect within the coating's microstructure was explained by the S-parameter and positron lifetime, obtained at a positron energy of 5 keV.

**RESULTS:** Figure 1 illustrates the variations in the S-parameter and the corresponding positron

lifetime of the two types of coatings, (a) H-DLC and (b) F-DLC, deposited at different bias voltages. For H-DLC coatings, the S-parameter and positron lifetime increased to ~0.492 and ~0.373, respectively, with increasing the negative voltage up to -5kV. However, the S-parameter decreased with a further increase in the negative bias voltage. This suggests that initially, the size of voids increased at -5 kV, and subsequently the number of  $\pi$  electrons increased. In the case of F-DLC coatings, the S parameter and the positron lifetime tended to increase simultaneously, but the change in the S parameter at -10 kV was not significant. This indicates that the size of voids monotonically increased with no significant difference in the number of  $\pi$  electrons at -10 kV.

- [1] A. Erdemir *et al.*, J. Phys. D Appl. Phys., **39** (2006) R311.
- [2] J. Choi *et al.*, Diam. Relat. Mater., **20** (2011) 845-848.
- [3] S. Nakao et al., Vacuum, 215 (2023) 112255.



Fig. 1. The changes in S-parameter and positron lifetime for (a) H-DLC and (b) F-DLC coatings as the negative bias voltage increases.

## Influence of Hydrogen Isotopes on Growth of Vacancy Clusters in Tungsten-Based Materials

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**INTRODUCTION:** As a plasma-facing material of a fusion reactor, tungsten (W) will be exposed to fuel particles, deuterium (D) and tritium (T), and products of fusion reactions, helium and 14 MeV neutrons. Vacancies and vacancy clusters formed by neutron irradiation act as strong traps against hydrogen isotopes and increase T inventory in W [1]. Understanding of vacancy formation and clustering is necessary for accurate evaluation of T inventory in a vacuum vessel of a fusion reactor. Displacements by neutrons occur under exposure to hydrogen isotope plasma, and hence hydrogen isotopes may affect on growth and decomposition of vacancy clusters.

The objective of this study is to investigate the influence of hydrogen isotopes on behaviors of vacancy and vacancy clusters to construct a kinetic model of clustering and annihilation. To reach this goal, W samples with monovacancies and vacancy clusters are prepared by electron beam irradiation. Then, the irradiated samples are heated with and without hydrogen isotopes, and the difference in size distributions of vacancy clusters is examined using positron annihilation spectroscopy.

**EXPERIMENTS:** Disk samples of W were wrapped with aluminum foils and immersed in a flowing water. Then the samples were irradiated with 8 MeV electrons using KURNS-LINAC to  $10^{-3}$  displacement per atom (dpa). A part of irradiated samples was exposed to D atoms at 200 °C to introduce D atoms and then heated under D<sub>2</sub> gas atmosphere at 0.1 MPa and 400 °C for 20 h. The other part of samples without D atoms were heated in vacuum at 400 °C for 20 h. Positron lifetime was measured by using <sup>22</sup>Na source at Institute for Materials Research, Tohoku University [1].

**RESULTS AND DISCUSSION:** The results of positron lifetime measurements are summarized in Table 1. The positron lifetime increased from 143 ps to 233 ps by irradiation of 8 MeV electrons. This means vacancy and vacancy clusters were generated by irradiation [2]. The exposure to D atoms resulted in reduction in positron lifetime to 204 ps because each trapped D atom brought 1 electron into a vacancy or vacancy cluster. Subsequent annealing at 400 °C under D<sub>2</sub> gas atmosphere led to increased positron lifetime of 226 ps because of growth of vacancy clusters. In contrast, annealing of the sample without D loading in vacuum at 400 °C resulted in reduction of lifetime to 212 ps due to decomposition of vacancy clusters. This clear difference indicated that vacancy clusters were stabilized and decomposition was hindered under the presence of D atoms.

### Table 1 Positron lifetime (ps)

Sample	As-received	e-beam irradiated	D atom exposed	Annealed under D <sub>2</sub> gas at 400 °C	Annealed in vac- uum at 400 °C
with D atoms	143	233	204	226	_
w/o D atoms	143	233	_	_	212

### **REFERENCES:**

[1] T. Toyama et al., J. Nucl. Mater., 499 (2018) 464-470.

[2] T. Troev et al., Nucl, Instr. And Meth. B, 267 (2009) 535-541.

## Impurity doping effect on vacancy-type defects in $\gamma$ -CuI studied using a slow positron beam

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**INTRODUCTION:** Copper iodide of  $\gamma$ -type ( $\gamma$ -CuI) is known as a promising candidate of the p-type semiconductor for thin-film transistors and hole transport layers in organic light emitting diodes (OLEDs). The effect of impurity doping on electronic properties of this material has been studied experimentally and theoretically [1]. It was recently reported that the hole concentration is controllable in the range of  $10^{14}$ - $10^{18}$  cm<sup>-3</sup> by zinc-doping [2]. This is based on the result that zinc doping reduces the hole concentration. The physical model for the reduction of hole concentration due to zinc doping has been proposed. One is that zinc atoms substituting copper atoms (Zncu) play as an electron donor. The other is that the zinc atoms introduce copper vacancyies (Vcu), leading to the formation of a Zncu-Vcu pair. To demonstrate which is more suitable model to explain the effect of zinc doping on electronic properties in zinc-doped  $\gamma$ -CuI, it is important to visualize the existence of Cu vacancies by positron annihilation spectroscopy (PAS). We performed positron annihilation lifetime spectroscopy (PALS) experiment of  $\gamma$ -CuI by using a slow positron beam from Kyoto University research reactor (KUR).

**EXPERIMENTS:** Samples were undoped and Zn and I-doped  $\gamma$ -CuI polycrystalline pellets. A Kapton film was used to evaluate the background of PALS spectra. PALS spectra of samples were measured at 300 K using a slow positron beam with the energy of 25 keV. The positron annihilation lifetimes (PALs) were analyzed using the software POSITRONFIT.

**RESULTS:** Figure 1 shows PALS spectra of undoped, Zn- and I-doped  $\gamma$ -CuI. The spectra are

reproduced by a single exponential decay component. The intensities reached over 95 %. The PALs were determined to be  $314\pm1$  ps,  $338\pm1$  ps and  $315\pm1$  ps for undoped, Zn- and I-doped  $\gamma$ -CuI, respectively. These values were almost agreement with the calculated PAL for the V<sub>Cu</sub> defect (=320 ps). It is apparent that the V<sub>Cu</sub> defect is dominant in our samples. This fact is reasonable, because the theoretical calculation of defect formation energies revealed that the V<sub>Cu</sub> defect is the most stable among vacancy-type defects in  $\gamma$ -CuI [2,3]. It was reported that Zn doping reduces the hole concentration. Actually, the reduction in hole concentration by Zn-doping was confirmed. However, it could not be clarified whether Zn-doping changes the concentration of V<sub>Cu</sub> defect. Further investigations are needed to clarify the effect of Zn-doping on the formation of V<sub>Cu</sub> defects in  $\gamma$ -CuI.



Fig. 1. PALS spectra of undoped, Zn- and I-doped  $\gamma$ -CuI, measured at 300K using a slow positron beam from KUR.

- [1] M. Grundmann et al., Phys. Status Solidi A, 210 (2013) 1671-1703.
- [2] M. Tsuji et al., ACS Appl. Mater. Interfaces, 14 (2022) 33462-33471.
- [3] K. Matsuzaki et al., J. Am. Chem. Soc., 144 (2022) 16572-16578.

## TDPAC Measurement of $^{111}Cd(\leftarrow^{111}In)$ in Ultrafine Bubble Water

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**INTRODUCTION:** Ultrafine bubbles, the gaseous cavities with diameters less than 1  $\mu$ m, have recently attracted much attention because of their multi-functionalities [1]. Fundamental studies on such multi-functionalities of ultrafine bubbles are not well extended because they are smaller than the wavelength of radiant rays. The time differential perturbed angular correlation (TDPAC) of <sup>111</sup>Cd( $\neg$ <sup>111</sup>In) in the aqueous solution with ultrafine bubbles is performed for the study of the interface of ultrafine bubbles, which should be one of the essential origins of its multi-functionality.

**EXPERIMENTS:** Typical four-counter TDPAC measurements were performed for the 171-245 keV cascade in <sup>111</sup>Cd( $\neg$  <sup>111</sup>In) in Oxygen water and water with Oxygen-ultrafine bubbles. The average diameter and the density of the ultrafine bubbles were 188 nm and 6.1 ×10<sup>5</sup>/mL, respectively. The aqueous solution of <sup>111</sup>InCl<sub>3</sub> at pH ~ 2 from Nihon Medi-Physics was added to each sample and adjusted to pH = 4.0 by just dilution with the respective solvent instead of adding NaOH or HCl in the previous studies [2][3]. The angular correlation term  $A_{22}G_{22}(t)$  is given by the following equation,

$$A_{22}G_{22}(t) = \frac{2(N(180^\circ, t) - N(90^\circ, t))}{N(180^\circ, t) + 2N(90^\circ, t)}$$

where  $N(90^\circ, t)$  and  $N(180^\circ, t)$  are the counting numbers of the 171-245 keV  $\gamma$ - $\gamma$  cascade at 90 and 180 degrees, respectively. The time-dependent term  $G_{22}(t)$  for each sample was obtained by normalizing  $A_{22}G_{22}(t)$  by the asymmetry parameter of 171-245 keV cascade in <sup>111</sup>Cd,  $A_{22} = -0.18$ .

**RESULTS:** Fig. 1 shows the observed  $G_{22}(t)$  for <sup>111</sup>Cd( $\neg$ <sup>111</sup>In) in Oxygen water and water with Oxygen-ultrafine bubbles. These were similar to the cases of  $G_{22}(t)$  in water at pH = 3.7 reported by Demille [3], but a slight difference was observed up to 60 ns be-



Fig. 2 TDPAC spectra of  $^{111}Cd(\neg ^{111}In)$  in ultrafine bubble water and Oxygen water at pH = 4.0.

tween these spectra. This difference may be caused by the influence of the ultrafine bubbles on the formation of hydrated ions of In. More studies, such as the detailed pH dependence, are underway.

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### **REFERENCES:**

[1] E. G. Denis, The fine bubble breakthrough. https://www.iso.org/news/2014/05/Ref1844.html

- [2] M. Tanigaki et al., KURNS Progress Report 2021, p.124.
- [3] M. Tanigaki et al., KURNS Progress Report 2022, p.123
- [4] G. R. Demille et al., Chemical Physics Letters, 44 (1976) 164-168.

## Dynamics Analysis of Helical Polymers Using Small Angle X-ray Scattering/Dynamic Light Scattering/Molecular Dynamics Simulation

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**INTRODUCTION:** Due to their potential applications in asymmetric catalysts, chiral stationary phases, and chiroptical materials, the structural control of helical polymers has received much attention. Recently, we have reported that helical poly(quinoxaline-2,3-diyl) (PQX) with chiral side chains display solvent-dependent helix inversions,<sup>1-3</sup> which can serve as effective frameworks for chirality-switching materials.<sup>4-7</sup> For elucidating the mechanism of helix induction and helix inversion of PQXs, we have studied the detailed structures of a PQX (100mer) with right- or left-handed structures into tetrahydrofuran- $d_8$  (THF- $d_8$ ) or a mixed solvent of 1,1,2-trichloroethane- $d_3$  (1,1,2-TCE- $d_3$ ) and THF- $d_8$  (4/1, v/v) using small-angle neutron scattering (SANS) experiments in conjunction with theoretical calculations.<sup>8</sup> Our recent interest has been focused on the effect of the dynamics of the side chains on the induction of the helix sense. Previously, we performed quasi-elastic neutron scattering (QENS) measurements to elucidate the influence of molecular dynamics on the solvent-dependent helix inversion of PQX. In this study, we performed molecular dynamics (MD) simulations to analyze the results of the QENS measurements in detail.

**EXPERIMENTS:** The initial geometries of the *P*- and *M*-helical PQX were estimated by SANS measurements in our previous study.<sup>8</sup> The MD simulations were performed using the Forcite module of Materials Studio 2020 with the COMPASS II force field (version 1.2) at ultra-fine quality. Here, we have treated the difference in viscosity between the two solvents as a difference in temperature. MD simulations of



Fig. 1 The initial structures of the PQX

the PQX were performed in a vacuum using a constant-volume/constant-temperature (NVT) canonical ensemble with a simulation time of 1000 ps at different temperatures (73.15 to 473.15 K).

**RESULTS:** We have compared the QENS simulations using the MD trajectories at varying temperatures and measured values. We found good agreement with the MD simulations at 313.15 K for the *P*-helical structure at 73.15 K for the *M*-helical structure. To elucidate the mechanism of helix inversion, we will analyze the obtained MD trajectories in the future.

- (1) T. Yamada et al., Chem. Commun., 46 (2010) 4914-4916.
- (2) Y. Nagata et al., J. Am. Chem. Soc., 135 (2013) 10104-10113.
- (3) Y. Nagata et al., J. Am. Chem. Soc., 137 (2015) 4070-4073.
- (4) Y. Nagata et al., Chem. Commun., 48 (2012) 11193-11195.
- (5) Y. Nagata et al., M. J. Am. Chem. Soc., 136 (2014) 9858-9861.
- (6) Y. Nagata et al., Angew. Chem. Int. Ed., 55 (2016) 7126-7130.
- (7) T. Nishikawa et al., ACS Macro Lett., 6 (2017) 431-435.
- (8) Y. Nagata et al., J. Am. Chem. Soc., 140 (2018) 2722-2726.

## Resonant frequency change with irradiation dose of piezoelectric PZT element

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**INTRODUCTION:** This study aims to establish an inexpensive method for dosimetry in high-dose environments. We consider changes in piezoelectric (PE) properties of PE elements due to irradiation for dosimetry in high-dose environments. For this purpose, the mechanism of radiation-induced change of PE properties has been investigated.

In a previous experiment conducted at NIRS/HIMAC, PZT PE elements were irradiated with 400 MeV/n Xe particles, and the decrease in the electromechanical coupling coefficient k was investigated [1][2]. As a result, it was found that  $k_r$  of the irradiated PZT element was -0.35%/kJ, which is a phenomenon concerning the irradiation dose [3]. To investigate what happens to PE elements due to irradiation, electron beam irradiation experiments have been conducted at KURNS-LINAC. The effect of temperature, which was negligible in the Xe particle experiments, has been investigated and controlled in the experiments at KURNS-LINAC [4].

**EXPERIMENTS IN THE FISCAL YEAR 2023:** In this study, to re-examine the effect of temperature rise on the piezoelectricity of PZT, PZT PE elements were irradiated with electron beams under sufficiently controlled beam intensity conditions to investigate the radiation-induced change in *k*. Furthermore, the retentivity of the change in *k* was verified by irradiating the device multiple times over a period of days [5]. The electron beam irradiation experiments were carried out twice, in June and December 2023. The irradiated samples were disc-shaped PZT (soft material, Honda Electronics) with a radius of 9 mm and thickness of 1 mm, and the irradiation experiments were carried out in the same way as before. The beam conditions were the same as before, with an energy of 20 MeV and a beam current (beam intensity) of  $1.1 \mu$ A, which was sufficiently low compared to the current value of 4.8  $\mu$ A in the previous experiment. The temperature of the controlled irradiated samples was constantly monitored during irradiation and air-cooling was used to ensure that the temperature did not rise above 60-70 °C. The temperature of the samples was monitored by thermocouples attached to the sample surface. The irradiation time per day was limited to about 2 hours, and irradiation was carried out over a period of 3-4 days. This was to determine the effect of the recovery of the *k*, which had changed while the irradiation was stopped.

**RESULTS:** The results of the two experiments were similar, except for the effects of poor measurements, which may have been due to inadequacies in the experimental set-up. The results can be summarized as follows. A change of more than one order of magnitude was obtained compared to the change in k due to temperature rise. This is the net k change due to electron beam irradiation. The result shows that the rate of change of k with respect to the absorbed dose (absorbed energy) applied to the PZT PE element, a value of  $-0.24 \pm 0.015$  [%/kJ] was obtained. [5]

### **REFERENCES:**

[1] M. Kobayashi et al., Jpn. J. Appl. Phys., 52 (2013) 126604.

[2] M. Kobayashi et al., Jpn. J. Appl. Phys., 53 (2014) 066602.

[3] S. Takechi et al., Jpn. J. Appl. Phys., 60 (2021) 038003.

[4] S. Takechi et al., Jpn. J. Appl. Phys., 61 (2022) 128001.

[5] M. Maeda, Master Thesis, Graduate School of Engineering, Osaka Metropolitan University (2024).

## Tritium release behavior from neutron irradiated LiAlO<sub>2</sub>

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**INTRODUCTION:** To launch a deuterium – tritium fusion reactor, it is required to produce enough among of tritium in advance. A high temperature gas-cooled reactor is proposed as a tritium production device [1]. One critical issue is tritium confinement at high temperature conditions. One idea is to combine alumina (Al<sub>2</sub>O<sub>3</sub>), which has a low hydrogen isotope permeability, with zirconium (Zr), which has a high hydrogen isotope solubility [2]. Tritium is generated in a lithium (Li) compound by a nuclear reaction with neutrons, and the tritium released from the Li compound is confined by the Al<sub>2</sub>O<sub>3</sub> wall. Zr packed within the Al<sub>2</sub>O<sub>3</sub> wall reduces tritium partial pressure and suppresses tritium permeation through Al<sub>2</sub>O<sub>3</sub> wall. LiAlO<sub>2</sub> is expected to be a promising Li compound due to its chemical stability. However, the tritium release characteristics from LiAlO<sub>2</sub> are not completely understood. In this study, Al<sub>2</sub>O<sub>3</sub> powder was irradiated by neutrons in Kyoto University Research Reactor and tritium release behavior by heating was observed.

**EXPERIMENTS:** Commercial powder of LiAlO<sub>2</sub> was packed in a quartz tube with vacuum sealing and was irradiated by neutrons with  $5.5 \times 10^{12}$  1/(cm<sup>2</sup> · s) in flux and 3 minutes in irradiation period. The weight of powder was 0.25 g. After neutron irradiation, the samples were transported to Kyushu University for tritium release experiments. The schematic diagram of the experimental apparatus is shown in Fig.1. The irradiated LiAlO<sub>2</sub> powder was packed in a quartz reaction tube. The sample was heated by an electric furnace to 900 °C in Ar gas flow. The released gaseous tritium (HT) and tritiated water vapor (HTO) were separately collected in water bubblers.

\*\*\*\*

**RESULTS:** Fig.2 shows the release behavior of tritium from neutron irradiated  $\text{LiAlO}_2$  powder. The chemical form of the released tritium was mostly tritiated HTO. HTO was released over a wide range of temperatures from 100 °C to 900 °C, with large peaks observed at around 250, 550, and 700 °C. The total amount of tritium released was  $1.0 \times 10^5$  Bq, and the ratio of HTO was 97 %, and the ratio of HT was 3 %. It was found that the tritium absorption performance of Zr from HTO and the permeation property of tritium from HTO through LiAlO<sub>2</sub> wall are important.

### **REFERENCES:**

[1]H. Matsuura, *et al.*, Nucl. Eng. Des., **243** (2012) 95–101.
[2]H. Nakaya, *et al.*, Nucl. Eng. Des., **292** (2015) 277–282.







Fig. 2. Tritium release behavior from neutron irradiated LiAlO<sub>2</sub> powder.

## Vacancy Migration Energy in VNbMoTaW High-Entropy Alloy

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**INTRODUCTION:** Tsai *et al.* [1] originally proposed the concept of sluggish diffusion in the high-entropy alloy(HEA). However, the reason for the sluggishness of diffusion is not quite clear. VNbMoTaW is known to form a quinary single-phased body-centered cubic structured solid solution. In the VNbMoTaW HEA at high temperatures, atomic diffusion is expected to proceed via a vacancy mechanism because they are substitutional solid solutions. Therefore, vacancy formation and migration energies in the VNbMoTaW HEA and its subsystems are important indexes for understanding the sluggish diffusion. In this work we have evaluated the vacancy migration energy in VNbMoTaW HEA by observing the vacancy migration and annihilation behavior during an annealing process after electron irradiation, with the use of the positron lifetime spectroscopy.

**EXPERIMENTS:** An ingot of VNbMoTaW HEA was cut into 10 mm  $\times$  10 mm  $\times$  1 mm plates, and carried out with solid solution heat treatment at 1423K for 24 h under argon atmosphere. The X-ray diffraction analysis shows that all the samples are composed of single phase with a bcc structure. Then, the samples were irradiated in water with 8 MeV electrons at a fluence of approximately,  $1.78 \times 10^{19}$  e<sup>-</sup> m<sup>-2</sup> below 373 K using the electron linear accelerator at the Institute for Integrated Radiation and Nuclear Science, Kyoto University. The irradiated samples were isochronally annealed in a temperature range from 373 to 1023 K. The temperature step during the isochronal annealing was 50 K and the duration of exposure to each temperature was 1 h. The positron lifetime measurements were made at 297–299 K.

**RESULTS:** Before the electron irradiation the positron lifetime spectrum for the solution-treated alloys was represented by only one component of 125 ps, which indicates that positrons annihilate in the bulk. After electron irradiation, the mean positron lifetime was increased to 155 ps. The analysis of positron lifetime spectra for the as-irradiated sample shows that many positrons are trapped and annihilate in the monovacancies introduced by electron irradiation, because the lifetime component,  $\tau_2$ , of trapped positrons was 176.7 ps. Fig.1 shows the relative change in the vacancy concentration during the isochronal annealing of the irradiated sample, which was evaluated on the basis of the two- or three-component analyses for the positron



Fig. 1. The relative change in vacancy concentration during the isochronal annealing of irradiated sample.

lifetime spectra. As shown in Fig.1, the theoretical curve of Dryzek *et al.*'s model [2] assuming that vacancy migration energy is 1.32 eV is in good agreement with the experimental data. The melting point of VNbMoTaW is reported to be 2946K. Taking into account the high melting point, the results of this work do not fully support the sluggish diffusion mechanism that has been proposed for HEAs.

### **REFERENCES:**

[1] K. Y. Tsai, M. H. Tsai and J. W. Yeh, Acta Mater., 61 (2013) 4887.
[2] J. Dryzek, C. Wesseling, E. Dryzek and B. Cleff, Mater. Lett., 21 (1994) 209.

## **Operando Measurement of a Lithium-Ion Battery Using an Internal Positron Source Technique**

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**INTRODUCTION:** Positron annihilation spectroscopy enables nondestructive detection of changes in electronic states of materials. However, in general, it is necessary to inject positrons from outside of the sample, making it difficult to investigate the interior of thick samples. For example, positrons emitted from the commonly used <sup>22</sup>Na positron source are injected only to a depth of less than 100 µm below the iron surface. Another technique is to inject a high-energy gamma-ray beam into the sample to pair-produce positrons inside the sample [1], but in this case it is not possible to investigate a specific region inside the sample. In this study, a lithium-ion battery (LIB) sample was exposed to thermal neutrons at KUR CN-3 to produce positron-emitting nuclide <sup>64</sup>Cu in the copper foil, which is the anode current collector. Then, the Doppler broadening of the annihilation radiation (DBAR) was measured while discharging the LIB sample.

**EXPERIMENTS:** Positrons emitted from the copper foil are injected into the graphite coated on the copper foil as an anode material. DBAR spectra were measured while discharging the LIB sample from a charged state over 25 hours using a fixed resistor circuit. The shape change of each spectrum was evaluated by the S-parameter [2].

**RESULTS:** Figure 1 shows the change in S-parameter caused by intercalation of lithium



Fig. 1. Dependence of S-parameters of graphite anode on Li concentration  $(Li_xC_6)$ .



Fig. 2. S-parameter change while discharging.

atoms into the graphite anode, obtained from first-principles calculations.  $Li_1C_6$  and  $Li_0C_6$  correspond to the charged and discharged states, respectively. During the discharge process of the LIB, the calculated S-parameters of graphite once increase and then begin to decrease. Figure 2 shows the change in the S-parameter measured while discharging the LIB sample. Although it should be noted that the discharge current was not constant (i.e., the Li de-intercalation rate was not constant), a trend like the change expected from the first-principles calculated and experimental values is mainly due to the use of different S-parameter definition formulas. We successfully probed the change of Li intercalation state in the anode of the LIB using positrons.

### **REFERENCES:**

[1] Y. Taira et al., Rev. Sci. Instrum., 93 (2022) 113304.

[2] F. A. Selim, Mater. Charact., **174** (2021) 110952.

## Vacancies in P-Doped β-FeSi<sub>2</sub> Film Probed By a Slow Positron Beam

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**INTRODUCTION:** As an environmentally friendly infrared luminescent material,  $\beta$ -FeSi<sub>2</sub> is expected. For practical use, however, a further increase in luminescence intensity is desired. Atomic vacancies have a significant impact on the luminescence characteristics in semiconductors. Using the KUR slow positron beam, vacancies in  $\beta$ -FeSi<sub>2</sub> films doped with Mn or Al, which are acceptor impurities that substitute the Fe and Si sites, respectively, have been investigated. As a result, Al doping, which has been reported to enhance luminescence intensity, was found to promote the formation of Fe vacancies. In this study, vacancies in  $\beta$ -FeSi<sub>2</sub> films doped with Co or P, which are donor impurities that substitute the Fe and Si sites, respectively [1,2], were studied.

**EXPERIMENTS:** The ion beam synthesis method was used to prepare undoped, Co-doped, and P-doped  $\beta$ -FeSi<sub>2</sub> films. These films were probed by a slow positron beam at the Kyoto University Research Reactor [3]. The Doppler broadening of annihilation radiation (DBAR) spectra were acquired. The shape of the obtained DBAR spectra was characterized by the S parameter [4], which depends on the fraction of positron annihilation with low-momentum electrons. Positrons trapped in vacancies increase the probability of annihilation of low-momentum valence electrons, resulting in an increase in the S parameter.

**RESULTS:** Figs. 1 and 2 show the S parameters of each film as a function of incident positron energy. The incident positron energy of around



Fig. 1. S parameters of undoped and Co-doped films as a function of incident positron energy.



Fig. 2. S parameters of undoped and P-doped films as a function of incident positron energy.

3-5 keV corresponds to the annihilation gamma-ray signal from the  $\beta$ -FeSi<sub>2</sub> film formed on the Si substrate surface. Focusing on that energy range, the S parameter is slightly lower in the Co-doped film, which substitutes Fe sites, than in the undoped film. In contrast, the S parameter of P-doped film, which substitutes Si sites, increases compared to the undoped film. This indicates that P-doping promotes vacancy formation. In the future, coincidence DBAR spectra will be measured to reveal whether the vacancies formed by the P-doping are Fe vacancies or Si vacancies.

### **REFERENCES:**

[1] J. Tani and H. Kido, J. Solid State Chem., 163 (2002) 248.

- [2] Y. Wanjun et al., J. Semicond., 34 (2013) 103003.
- [3] A. Yabuuchi, Nucl. Instrum. Method Phys. Res., Sect. B, 513 (2022) 44.
- [4] F. Tuomisto and I. Makkonen, Rev. Mod. Phys., 85 (2013) 1583.

## **Radiation Induced Demagnetization of Neodymium Magnets**

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**INTRODUCTION:** Permanent magnets are essential devices in the transport and control of charged particle and neutron beams due to their lower power consumption, compactness, and the fact that they do not require ancillary equipment [1-3]. On the other hand, demagnetization due to radiation is one of the concerns when using permanent magnets, but quantitative evaluation of its effects is insufficient. Therefore, we have systematically evaluated the effects of radiation-induced demagnetization using the irradiation facility at KUR over the last few years [4]. Magnet manufacturers add several additives to improve coercivity. Naturally, the additives are trade secrets and undisclosed. Previous studies have shown that even the same neodymium magnets show significant differences in the rate of demagnetization caused by radiation, and one of the reasons for this is thought to be the dysprosium content. However, other substances are also expected to have a significant influence on resistance to radiation. As additives other than the main magnet component have not been published, an attempt has been made to measure the correlation between additives and the effect of demagnetization.

**EXPERIMENTS:** In this study, differences in radiation demagnetization effects for neodymium magnets were evaluated. The irradiated sample were five types of neodymium magnets; N38H, N38EH, N39SH, N39UH, N48H made by SHIMONISHI SEISAKUSHO Co,. Ltd. The shape of the sample was cylindrical with a thickness of 1 mm and a diameter of 5 mm. The irradiation experiments were performed at KUR Pn-2 port and Tc-Pn. Irradiation was also carried out for elemental identification using activation analysis methods.

**RESULTS:** The evaluation of demagnetization is ongoing, as we need to wait for the radiation levels of the samples to decrease. Fig. 1 shows the typical gamma-ray spectrum taken with a Ge detector at about 3.5 hours after irradiation of 3 seconds using Pn-2. As shown in Fig. 1, it is expected that there are differences in the substances contained in N38H and N38EH as well. Measurements with a Ge detector



Fig. 1. Gamma-ray energy spectrum taken with a Ge detector at about 3.5 hours after irradiation of 3 seconds using Pn-2. Upper is N38H, lower is N38EH, respectively.

have been made at several times after irradiation. The nuclides will be identified and their correlation with radiation demagnetization will be investigated in detail.

- [1] Y. Fuwaet al., Prog. Theor. Exp. Phys., 2017(2) (2017) 023G01.
- [2] Y. Kuriyamaet allEEE Trans. Appl. Supercond., 34(5) (2024) 4004005.
- [3] M. Yamada et.al., Prog. Theor. Exp. Phys., 2015(4) (2015) 043G01.
- [4] Y. Fuwa et al., MT-27 Int. Conf. on Magnet Technology, (2021) TUE-P01-115-05.

## Performance of Field Emitter Based Photo-detection Device under Gamma-ray Irradiation

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**INTRODUCTION:** One of the most important issues for nuclear decommissioning of Fukushima Daiichi nuclear power plant is to develop image sensor reliable even in the radiation circumstance. The authors have been developing an image sensor based on the field emitter arrays, exhibiting the high durability against  $\gamma$ -ray irradiation [1]. We have already shown the fundamental characteristics of the photo-detection device based on field emitter array (FEA) under a normal circumstances [2]. In this study, the operational performance of the photo-detection device under  $\gamma$ -ray irradiation and in normal circumstances were investigated.

**EXPERIMENTS:** The photo-detection device was consisted of an FEA, mesh electrode, and photoconductor. The FEA which was used in the previous study [2] was again used.  $\gamma$ -ray irradiation was performed at the Co-60 gamma-ray irradiation facility, Research Institute for Integrated Radiation and Nuclear Science, Kyoto University. The emitter was grounded and the gate was biased to 60 V to extract electrons. The mesh was given a potential of 200 V. The anode characteristics was acquired with varying the anode voltage from 0 V to 50 V. The dose rate of irradiation was approximately 50 – 60 Gy h<sup>-1</sup>.

**RESULTS:** The signals of visible light and  $\gamma$ -ray are shown in Fig. 1 [3]. The dotted line shows the net  $\gamma$ -ray signal which was obtained by subtracting the dark current from the anode current under  $\gamma$ -ray irradiation. The solid



Figure 1. Anode characteristics under visible light and  $\gamma$ -ray irradiation.

line shows the net visible light which was obtained by subtracting the dark current from the anode current under visible light irradiation. The power of the light was 10  $\mu$ W. It is clearly seen that the anode current flows only a little under the  $\gamma$ -ray irradiation, as compared to the current with visible light incident. It is clearly seen that the present device has higher sensitivity for visible light than for  $\gamma$ -ray. From this result, the device could be used as a in the primary containment vessel in which the dose rate is approximately 100 Gy h<sup>-1</sup>.

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### **REFERENCES:**

[1] Y. Gotoh et al., IEEE Trans. On ED 67 (2020) 1660-1667.

- [2] Y. Gotoh et al., KURNS Progress Report 2022 (2023) 114.
- [3] Y. Gotoh et al., The 58<sup>th</sup> Meeting of KURNS 2024, P15.

### Fundamental study of damage on tungsten by heat and particle loading

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**INTRODUCTION:** It is of a great importance to clarify phenomena of implantation, retention, diffusion and permeation of tritium (T) on surface of the armor materials of the first wall/blanket and the divertor on the fusion devices from a viewpoint of precise control of fuel particles, reduction of tritium inventory and safe waste management of materials contaminated with tritium. In the present works, T exposure experiments have been carried out on W samples which were irradiated by high energy electrons to investigate effects of high energy electrons irradiation on microstructure and tritium retention of W. In this fiscal year, pure W and recrystallized W were irradiated by high energy electron beam. Before and after that, positron annihilation experiment was carried out to identify the radiation defect. In this fiscal year, tritium exposure experiments at 350 °C using a tritium (T) exposure device is reported.

**EXPERIMENTS:** W samples used were ITER specification W (ALMT-grade) (SR-W) and its recrystallized W (RC-W). The SR-W was fabricated via a powder metallurgical route including cold isostatic pressing, sintering, hot rolling, and heat treating to relieve the residual stresses. Some of the machined SR specimens were subjected to a full recrystallization treatment at 2000 °C for 1 hr in vacuum. High energy electrons irradiation has been carried out using LINAC in Institute for Integrated Radiation and Nuclear Science, Kyoto University. An peak energy of electron irradiated was 8 MeV and DPA was 5.8 x 10<sup>-3</sup>. Temperature during the irradiation was measured by thermocouples which was contacted with a backside of the W samples. Before and after that, positron annihilation experiment was carried out to identify the radiation defect. In addition, a high energy ion irradiation experiment has been carried out. The sample surface was irradiated by 2.5 MeV Fe ions with a flence of 5 x  $10^{18}$  ions/m<sup>2</sup> at RT. T exposure experiments have been carried out using a T exposure device in University of Toyama. Pressure of the T gas was 1.3 kPa and T exposure was kept for 4 h at 350 °C. T concentration in the gas was about 5 %. After the exposure to T gas, T amount retained in surface layers of the sample was evaluated by imaging plate (IP) measurements and  $\beta$ -ray-induced X-ray spectrometry (BIXS).

**RESULTS:** Figure 1 shows X-ray spectra observed by BIXS. Signal of  $Ar(K_{\alpha})$  is from T near the sample surface. On the other hand,  $W(M_{\alpha})$  shows amount of T near a depth of a few µm from the surface. The results indicate that depth distributions of T are different between e-irradiated and Fe irradiated samples because the ratios of  $W(W(M_{\alpha}))/Ar(K_{\alpha})$ are different. These results are considered to be related with T retention by radiation damage near the sample surface.



Fig. 1. Results of BIXS measurement. T exposure temperature is 350 °C. (a): SR-W, (b): electron irradiated SR-W, (c): Fe irradiated SR-W, (d): RC-W, (e): Fe irradiated RC-W, (f): Fe irradiated RC-W.

### Correlation between irradiation defect density and hydrogen isotope retention in tungsten

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**INTRODUCTION:** Tungsten (W) will be used in the deuterium-tritium (D-T) fusion reactor to face to energetic particles such as fast neutrons. W atoms at lattice points in the crystal structure are recoiled by fast neutrons, forming irradiation defects. About two orders of magnitude larger D retention in neutron irradiated W has been reported [1]. This result should suggest that irradiation defects strongly trap hydrogen isotopes, leading to a drastic change of hydrogen isotope transport behaviors in W. In this study, we tried to evaluate the quantitative correlation between irradiation defect density and hydrogen isotope retention in W.

**EXPERIMENTS:** The W sample was purchased from A.L.M.T co. The thickness of the sample was 500 µm. A sample was pre-annealed at 1173 K for 0.5 hours under high vacuum. Then, samples were shipped to KURNS-LINAC for electron irradiation. The electron acceleration voltage was 8

MV. The dose was evaluated as  $4.3 \times 10^{-3}$  dpa according to an atomic displacement cross-section of 70.4 barns and a displacement threshold energy of 84 eV [2]. The irradiation temperature was less than 353 K due to the water coolant.

For evaluating the irradiation defects formed in the sample, positron lifetime measurements were conducted at Kagoshima Univ. The sample was faced to <sup>22</sup>Na deposited on the plastic substrate. <sup>22</sup>Na emits a positron and a 1.27 MeV gamma-ray. The scintillators surrounding the sample detected 1.27 MeV gamma-ray, then the elapsed time to detect the posi-

tron-electron annihilation gamma-rays was measured as the positron lifetime.

**RESULTS:** The results of the positron lifetime measurements are shown in Fig. 1. The results of annealed W and electron irradiated W were similar, although a slight difference can be found in the decay behaviors. From the decay behaviors the positron lifetimes of annealed W and electron irradiated W were evaluated to be 151 ps and 186 ps, respectively. The components with longer positron lifetime were not detected. This indicates that small size defects such as mono-vacancies and dislocation loops were formed by electron irradiation.

To measure the trapping behavior of hydrogen isotopes in defects produced by electron irradiation, tritium ion irradiation was carried out using the tritium ion irradiation apparatus shown in Fig. 2 placed in Hydrogen Isotope Research Center, University of Toyama. The depth profile of tritium concentration will be measured by the combination of electrolytic polishing and the liquid scintillation counter. For these measurements, the polishing rates of W materials are estimating by the weight loss method, and the tritium concentration measurement in electron irradiated W will be done in FY2024.

### **REFERENCES:**

[1] Y. Hatano *et al.*, Nucl. Fusion, **53** (2013) 073006.
[2] K. Sato *et al.*, Nucl. Mater. Ene., **9** (2016) 554.



Fig. 1 The results of positron lifetime measurements.



Fig. 2 Tritium ion irradiation apparatus

## Neutron Irradiation Tests for Components of ITER Diagnostics System

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**INTRODUCTION:** ITER [1] is the world's largest fusion experiment reactor and is under construction in southern France. Components of diagnostic systems installed in the ITER tokamak are exposed to high neutron radiations. Therefore, it is important to investigate the effect of neutron irradiation on these components. In this study, neutron irradiation tests on many kinds of components of diagnostic systems of ITER have been conducted. This report presents the result of the neutron irradiation test on the ultrasonic motor of Infrared Thermography system (IRTh) [2] and the pressure gauge of the Edge Thomson Scattering system (ETS) [3] and Divertor Impurity Monitor system (DIM) [4].

### (1) Irradiation test for the ultrasonic motor for IRTh

IRTh plans to use a steering mirror driven by an ultrasonic motor for optical axis adjustment in the ITER. In this study, neutron irradiation tests on the ultrasonic motor were conducted.

**EXPERIMENTS:** The ultrasonic motor was irradiated at neutron fluence up to  $1.0 \times 10^{16}$  (n/cm2), equivalent to neutron fluences during 20 years of ITER operations. The characteristics of rotational speed relative to torque for the motor were compared before and after the irradiation under three conditions: when the torque was zero (no load) at rotational speeds of 80, 120, and 150 rpm.

**RESULTS:** Figure 1 shows the irradiation test result of the ultrasonic motor. As shown in Fig. 1, a slight degradation in torque was observed due to the neutron irradiation up. However, since the torque required for driving the steering mirror is approximately 0.2 Nm, focusing around 0.2 Nm revealed no significant performance degradation was observed.



Fig. 1: Rotational speed relative to torque for the ultrasonic motor before and after neutron irradiation

### (2) Irradiation Tests of the pressure gauge of DIM, ETS and IRTh

DIM, ETS and IRTh plan to use pressure gauges in the port cell area of ITER. In order to estimate how much neutron shielding is needed to use the pressure gauge without replacement during 20 year's ITER operation, the neutron irradiation tests on the pressure gauge were conducted.

**EXPERIMENTS:** For the irradiation test of the pressure gauge, the Slant exposure tube was used. The pressure gauge was irradiated at multiple neutron fluences up to  $2.0 \times 10^{12} \text{ n/cm}^2$ . The performance of the pressure gauge was compared before and after the neutron irradiation.

**RESULTS:** No change in the performance of the pressure gauge was observed before and after the irradiation up to  $2.0 \times 10^{12} \text{ n/cm}^2$ . The neutron irradiation test will be continued at neutron fluence up to  $1.0 \times 10^{13} \text{ n/cm}^2$  which is equivalent to 20 years of ITER operation to investigate the effects of neutrons.

### **REFERENCES:**

[1] B. Bigot, Fusion Eng. Des., 146 (2019) 124.

[2] T. Ushiki et. al., Fusion. Eng. Des., 168 (2021) 112665.

- [3] E. Yatsuka et. al., Fusion. Eng. Des., **136** (2018) 1068.
- [4] H. Ogawa et. al., Kasai, Fusion Eng. Des., 83 (2008) 1405.

## **Electron Microscopy of Pure Aluminium Deformed at Static High Pressures**

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**INTRODUCTION:** Metals are often processed to experience severe plastic deformation to modify their physical properties. A fine-grained microstructure with grain sizes of several hundreds of nanometers or less often forms after such processing. It has been known that, after extensive deformation processing, metals without alloying elements, such as pure aluminum, can significantly increase their physical strength. This is considered to be primarily because of their grain size reduction. In addition to the grain sizes, dislocation density, and lattice-preferred orientation are considered to be important features that physically affect the strength of metals. Therefore, it is important to transform such microtextural features of pure metal and analyze them using nano-scale analytical methods.

EXPERIMENTS: We have been trying to transform and analyze microstructures of pure aluminum experiencing extensive deformation at controlled static high-pressures using a diamond anvil cell apparatus, which is capable of generating pressures of the order of 10 GPa  $\sim 10^5$  atmospheres [1,2]. Aluminum powder with a grain size of 75 µm and purity of 99.99% was annealed at 250°C for 24 hours to prepare initial specimens. This annealing procedure effectively removed dislocations. The specimens were then packed and compressed in a sample chamber of a diamond anvil cell ap-paratus of 600 µm culet size with a metal plate gasket and without a pressure medium to induce a high-strain environment at high sealing pressure. The pressure was determined by the ruby fluores-cence method. The recovered specimens were cut and thinned by focused ion beam apparatus, and the thin sections were observed by transmission electron microscopy to analyze inter-grain orienta-tion differences, dislocation densities, and grain size trends [1,2].

**RESULTS:** A typical result of transmission electron microscopy is shown in Fig. 1, which was taken using a bright-field method. The average grain sizes were determined by image analysis of these results. Orientations of the observed crystals grins were determined using the limited-field electron diffraction method, and the orientation angle differences between the grains were measured. It was found that the average orientation difference between grains increased with increasing pressure [1]. As this finding, the microstructures of aluminum obtained after extensive compression and deformation at static high pressures showed some systematic characteristics [2], which will be summarized Fig. 1. Aluminum after compression at 1.5 GPa and reported separately.



- [1] Y. Tanaka et al., Special Issue of the Re-view of High Pressure Science and Technology, 31 (2021) p.104 (1P04).
- [2] T. Okuchi et al., S 2023 Annual Meeting of Japan Associaton of Mineralogical Sciences Abstract Volume, R3-13.

## Study to improve transport and measurement performance of a slow positron beamline

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**INTRODUCTION:** Positron annihilation spectroscopy is an important analytical method to detect vacancy-type defects and vacant spaces of materials. Energy-variable mono-energetic positron beams (slow positron beams) are essential to perform depth-dependent positron annihilation spectroscopy of surface layers such as ion-implanted layers or thin films formed on substrates. As moderation efficiencies to obtain slow positron beams are not very high, intense positron sources are required for practical use. In general, positron sources based on pair creation can provide higher intensity than radioisotope-based positron sources. A positron source using pair-creation by gamma-rays from a nuclear reactor have been developed by using Kyoto University research Reactor (KUR) to obtain a slow positron beam for materials analysis. In the KUR slow positron beam-line, a lifetime measurement system based on a pulsing system using a radiofrequency-driven buncher was developed for the KUR slow positron beamline. In this report, standard samples for positron annihilation lifetime spectroscopy using a slow positron beam were prepared to obtain better performance of the positron lifetime system.

**EXPERIMENTS:** Standard samples for the lifetime of Si divacancies were prepared by 200 keV Si ion irradiation at room temperature to FZ-grown Si samples of  $1.5 \times 1.5$  cm. The amorphization threshold dose for 200 keV ion irradiation to Si was estimated to be around  $2 \times 10^{14}$  cm<sup>-2</sup> [1]. Thus, several irradiation doses in the range of  $2 \times 10^{12} - 2 \times 10^{14}$  cm<sup>-2</sup> were examined with respect to this threshold dose. Positron annihilation lifetimes of the irradiated samples were measured by the KUR slow positron beamline at an incident energy of 6 keV.

**RESULTS:** Fig. 1 shows the positron annihilation lifetime spectra of Si samples irradiation to dose of  $2 \times 10^{12}$  and  $2 \times 10^{13}$ cm<sup>-2</sup> compared with a Kapton spectrum. The obtained lifetime spectra of the Si-irradiated samples were analyzed by the PALSfit code. The  $2 \times 10^{13}$  sample gave one lifetime component of  $\tau = 0.3037 \pm 0.007$  ns. The measured value is in agreement with the reported Si-divacancy lifetime [2]. The  $2 \times 10^{12}$ sample gave two lifetime components of  $\tau_1$ =0.0271±0.0137 ns  $\tau_2$ =0.3011±0.0011 ns, where the annihilation in bulk was detected. Considering about the easiness of the sample preparation and lifetime measurement, the  $2 \times 10^{13}$ sample is better as a standard sample. In particular, unirradiated Si cannot be used as a standard for positron energies below 20 keV owing to the positron diffusion to the surface. The standard samples prepared in this study is useful.

### **REFERENCES:**

[1] A. Kinomura *et al.*, Appl. Phys. Lett., **88** (2006) 241921.
[2] G. Amarendra *et al.*, Phys. Rev. B, **63** (2001) 22411.



Fig. 1. Positron lifetime spectra for Si-irradiated Si and Kapton samples.

### Development of a slow positron source using a electron linear accelerator

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**INTRODUCTION:** Slow positron beams (energy-variable monoenergetic positron beams) have been widely used to investigate vacancy-type defects in thin films or near-surface layers of various functional materials. Intense slow positron beams can be generated exclusively from large-scale experimental facilities such as nuclear reactors and accelerators, while radioisotope-based positron sources are used for medium intensity facilities. In terms of accelerator-based positron sources, so far, radiofrequency-driven electron linear accelerators (LINAC) have been used. In principle, LINAC-generated positron beams are not continuous (i.e., pulse beams), while reactor-based positron beams are continuous. The pulse beams can be potentially applied for unique applications such as pump-probe type measurements [1].

**EXPERIMENTS:** The KURNS-LINAC was used to generate slow positrons in this study. The KURNS-LINAC with two acceleration tubes driven by L-band (1.3 GHz) radiofrequency can generate 30 MeV electron beams with a repetition rate around 100 Hz and a pulse width up to 5  $\mu$ s. Fig. 1 shows the plan view image of the beamline configuration. It should be noted that preliminary experiments using the KURNS-LINAC reported slow-positron beam generation with intensities of >10<sup>7</sup> e<sup>+</sup>/s [2].

**RESULTS:** By using the SIMION code for charged-particle trajectory simulation, the performance of the conventional beamline components such as mesh-shaped moderators, magnetically guided beam ducts and a linear storage section, have been evaluated for effective beam generation and transport. As result of the SIMION simulation, cylindrical electrodes were used for a linear storage section to obtain higher positron intensity. The calculation using the PHITS code was used optimize the converter to generate positrons through pair creation reactions. Consequently, a tungsten converter with a thickness of 4 mm was used. Mesh-type moderator electrodes with annealed Pt foils with thickness of 50  $\mu$ m was installed. The calculation indicated that the slow positron intensity close to  $6 \times 10^6 \text{ e}^+/\text{s}$  can be expected for the current setup assuming a moderator efficiency of  $4.5 \times 10^{-5}$  [3]. Beamline components designed based on the conceptual design were made and installed in the target and measurement rooms of the KURNS-LINAC building as shown in Fig. 1.

Preliminary experiments to generate slow positron beams were performed using a 30 MeV electron beam. As the beamline construction was not completed, the generated slow positron beam was detected at the end of the linear storage section by a scintillation detector. Signals attributed to the slow positron beam were clearly detected. The storage section successfully trapped positron beams in the range close to 1 ms. Currently, the beamline was fully connected and extended to a sample chamber.



Fig. 1 Plan-view image of the slow positron beamline using the KURNS-LINAC.

### **REFERENCES:**

[1] A. Kinomura et al., Rev. Sci. Instrum., 85 (2014) 123110.

[2] H. Tanaka et al., Jpn. J. Appl. Phys., 31 (1992) 4029.

[3] B.E. O'Rourke et al., Rev. Sci. Instrum., 82 (2011) 063302.

## Effects of Pulse Irradiation by Charged Particles on Damage Structures in Metals

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**INTRODUCTION:** For the development of nuclear materials, irradiation experiments are important. High energy charged particles such as ions and electrons, which are produced in accelera-tors are frequently employed for these experiments. Ion and electron beams from accelerators driv-en by radiofrequency or accelerators with beam scanners possess time structures (period-

ic/discontinuous intensities). Examples of

pulse trains are shown in Fig. 1. The pulse frequency is 1/T and H is the beam intensity, but we consider H to be the damage rate (dpa/s) in a pulse duration  $\tau$ . It's uncertain if the irradiation effects of these pulse beam operations change depending on the pulse frequencies.



Fig. 1 Examples of time trains.

**EXPERIMENTS:** For the study of pulse frequency effect on damage structures, the electron irradiation experiments with two irradiation frequences, 10 Hz and 100 Hz, were performed by the KURNS-LINAC with an acceleration energy of 8 MeV and a pulse width of 5  $\mu$ s at 363 K to a dose of 2.8×10<sup>-4</sup> dpa. The positron annihilation lifetime spectroscopy (PALS) of the specimens was measured at room temperature using Na-22 and a conventional fast–fast spectrometer. The lifetime spectra were analyzed using the PALSfit program [1].

**RESULTS:** Table 1 shows the results of PALS in Ni and Fe. The long lifetimes 220 ps (100 Hz) and 240 ps (10 Hz) in Ni indicate the existence of micro-voids of 3 and 4 vacancies in average, respectively. In Fe, there exist micro-voids of 5 vacancies (100Hz) and 9 vacancies (10 Hz) [2]. These results clearly indicate the existence of irradiation frequency effects. For the understanding of the effects, the reaction kinetics analysis is in progress.

	Short lifetime	Intensity of short lifetime	Long lifetime	Intensity of long lifetime	Averale lifetime
Ni-100Hz	0.1094	86.6	0.2212	13.4	0.1244
	(±0.0018)	(±2.74)	(±0.0146)	(±2.72)	(±0.0011)
Ni-10Hz	0.1064	86.8	0.2412	13.2	0.1243
	(±0.0013)	(±1.67)	(±0.0115)	(±1.66)	(±0.0011)
Fe-100Hz	0.1047	79.2	0.2476	20.8	0.1344
	(±0.0014)	(±1.61)	(±0.0081)	(±1.58)	(±0.0052)
Fe-10Hz	0.1007	80.4	0.2802	19.6	0.1359
	(±0.0010)	(±0.877)	(±0.0061)	(±0.858)	(±0.0032)

Table 1. PALS of Ni and Fe irradiated at 363 K to  $2.8 \times 10^{-4}$  dpa.

### **REFERENCES:**

[1] P. Kirkegaard et al., Roskilde: Technical University of Denmark; 2009.

[2] H. Ohkubo al., Mater. Sci. Eng. A, 350 (2003) 95-101.

### Research on freezing of materials under high pressure by Bessel-beam driven micro-explosions in transparent materials

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**INTRODUCTION:** It is possible to create extreme pressure and temperature conditions in the tabletop laboratory experiments with 100-fs ultra-short laser pulses focused inside a transparent material, with intensity in the focal spot significantly above the threshold for optical breakdown [1]. Tight focusing of a -laser pulse with a few micro-Joule energy deep inside a crystal can deposit an energy density up to several MJ/cm<sup>3</sup> in a sub-micron volume. Pressure created by the plasma expansion is high enough to generate new material phases, which is referred to as micro-explosion [2,3]. Here we report an experiment relative to a micro-explosion. Targets were transparent oxide materials (MgO). Hence, we demonstrated X-ray diffraction (XRD) analysis of the material under high pressure and high temperature phase.

### **EXPERIMENTS:**

(1) Micro-explosions experiment with femtosecond laser

The beam of a 250 fs amplified Ti:Sapphire laser with a central wavelength of 800 nm was focused inside MgO crystal with depth of 30  $\mu$ m from the surface. The energy and spot size were 200 nJ and 1.2  $\mu$ m, respectively.After laser processing, the structures of void inside MgO were observed with Scanning Electron Microscope (SEM).

(2) XRD analysis with by Rigaku Ultima IV at KURNS

XRD analysis was demonstrated with the material with void formed inside by micro-explosions to observe high pressure phase of Mg. X-ray energy was 8.04 keV (Cu-Kα).

### **RESULTS:**

Results of observation of void structure with SEM shows the length and diameter were 30  $\mu$ m and 350 nm, respectively. The volume of the void is ~2 femtolitres.

Figure 1 shows the results of XRD. Signal of  $2\theta$  diffraction with angles of 44.5 and 64.45 degrees. These observations indicate the signals derived from Mg high pressure phase (Mg-bcc) [4]. Further analyses are underway to better understand the data

### **REFERENCES:**

S. Juodkazis *et al.*, Phys. Rev. Lett.,
 **96** (2006) 166101.
 A. Vailionis *et al.*, Nat. Com-mun. 2
 (2011) 445.
 L. Rapp *et al.*, Sci. Rep., 6 (2016)

34286. [4] C. W. Greeff and John A. Mo-riarty,

Phys. Rev. B, **59** (1999) 3427.



## Irradiation Damage Study of New Materials and Industry Materials Under Irradiation and after Irradiation

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**INTRODUCTION:** Materials and instruments used in a radiation environment must have irradiation resistant properties. Therefore, it is important to promote research and development not only of existing applied and industrially developed materials, but also of innovative materials such as high entropy alloys [1,2] with even better properties. One of the objectives of this study is to understand the basic process of irradiation damage of high-entropy alloys as innovative materials. The electron-beam irradiation method, which can produce the simplest lattice defect, the Frenkel pair (energy must be adjusted), is an important experimental technique. Our efforts in this study have another objective. This is to detect the appearance of elastic waves produced by the stresses to which the material is subjected by the pulsed beam, and to measure and evaluate them using the four-terminal electrical resistance method as a nondestructive testing technique.

**EXPERIMENTS:** The electron irradiation experiments were performed using the electron linear accelerator at Kyoto University. In order to investigate the threshold energy of the atomic displacement process in Fe- and Ti-based high-entropy alloys (Fe-Mn-Cr-V-Al-C and Ti-V-Cr-Zn-Ta) caused by collisions with energetic incident particles, the temperature of the samples was kept at temperature about 4.0 K in a vacuum and the acceleration voltage of the electron beam was varied from 0.3 MV to 2.0 MV, and the electrical resistance of the sample was measured at each acceleration voltage by the four-terminal electrical resistance method [3]. Elastic waves excited in the solid by pulsed electron beam waves (10, 20, and 30 Hz) around room temperature were also measured by the same 4-terminal electrical resistivity method in air at an energy of 8.0 MeV.

**RESULTS:** While the electrical resistance of Fe-based HEA began to change at about 0.25 MeV to 0.3 MeV, that of Ti-based HEA was found to change for the first time at about 1.8 MeV, which is considered higher than the generally assumed threshold energy of Ti alloys. On the other hand, the results of the measurement by the 4-terminal electrical resistance method for elastic waves (stress waves) in solids induced by pulsed electron beams in the temperature region above room temperature were as follows. In 316L steel, the change in electrical resistance increased with increasing frequency (10-30 Hz); in Fe-based HEA, the change was significantly larger than that in annealed 316L steel, which was assumed to be due to the interaction between the elastic waves and the unique internal strain of the HEA alloy. The results of this research and a detailed investigation of these material properties will be submitted to journals in the near future [4,5].

### **REFERENCES:**

[1] E. Wakai et al., Mater. Char., 211 (2024)113881.

- [2] E. Wakai et al., Sci. Talks, 8 (2023)100278.
- [3] Y. Iwamoto et al., J. Nucl. Mater., 458 (2015) 369-375.
- [4] E. Wakai et al., Nature (or Nature Mater. or Nature Com.) to be submitted in 2024.
- [5] E. Wakai et al., Journal to be submitted in 2024.

## Attempt at neutron activation analysis of wood using the k<sub>0</sub>-method

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**INTRODUCTION:** The Space-wood project (LignoStella Project), which is conducted by Kyoto University in collaboration with Sumitomo Forestry Co. Ltd., aims to explore the use of wood materials in outer space. The project is currently underway and plans to launch a small wooden satellite in 2024. Researchers are particularly interested in understanding the effects of cosmic radiation on wood. Secondary cosmic rays, specifically neutrons, are generated when protons from cosmic radiation collide with the International Space Station (ISS) vehicle. There has been few studies of the effects of neutron collision on wood chemical components [1]. In this study, we attempt to analyze the neutron activation of wood using the  $k_0$ -method [2].

**EXPERIMENTS:** Three types of specimens were cut from a honoki board (*Magnolia obovata*): sapwood, intermediate wood, and heartwood. Each specimen had dimensions of 75 mm (length) × 10 mm (radial) × 10 mm (tangential). After measuring the dimensions and weight, the specimens were stored in an air-dry state at room temperature. The test specimen was irradiated with neutrons using KUR's pneumatic transport tube (pn-3) for 60 seconds (1 MW). Following neutron irradiation, the test specimen underwent gamma ray spectrometry using a Ge semiconductor detector immediately, 1 hour later, and 24 hours later. By analyzing the measured  $\gamma$ -ray energy spectrum, the k0 method was employed on a computer to identify and quantify the elements.

**RESULTS:** As shown in Fig. 1, K, Mg, Cl, Na, Ar, Mn, Al, and Sr were confirmed as trace elements that become  $\gamma$ -ray producing nuclides upon activation in Honoki. Concentration changes in the trunk differed depending on the element, with some concentrations being higher in the sapwood (K, Cl), some in the heartwood (Ar, Al), and some with less clear change trends (Na, Mn). In addition, Mg increased from sapwood to intermediate wood, but was not detected in heartwood in all individuals. It was found that the identification and quantification of trace elements within an individual is possible using the k<sub>0</sub> method.



Fig. 1. Nuclides detected by  $K_0$  method in honoki. A, B and C are sapwood, intermediate wood and heartwood, respectively.

### **REFERENCES:**

[1] Y. Katayama et al., Radioisotopes, 35(11) (1986) 577-582.

[2] Moens L. et al., Journal of Radioanalytical and Nuclear Chemistry, 82(2) (1984) 385-452.