# Artificial Alpha Recoil Track Formation on Synthetic Zircon: Preliminary Observation

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**INTRODUCTION:** <sup>238</sup>U, <sup>235</sup>U, and <sup>232</sup>Th decay through emission of alpha particles to stable lead. Alpha Recoil Track (ART) is the damage formed when daughter nuclides move back in responce to alpha decay (Fleisher, 2003). Given the known decay constants of uranium and thorium, the age since the start of ART accumulation can be calculated by measuring the number of ARTs and the uranium and thorium concentrations. Zircon is rich in uranium and thorium, and is often analysed in geochronological studies. Unlike the layered silicates such as muscovite or biotite, ART in zircon has never been observed before, despite its potential to date samples whose ages are in order of thousand years old or older.

The purpose of this research is to establish a method to identify the ARTs in zircon by using the synthetic zircon and the artificial formation of ARTs using <sup>241</sup>Am source.

**EXPERIMENTS:** Synthetic zircon was formed by the flux method from the reagents of ZrO<sub>2</sub>, MoO<sub>3</sub>, Li<sub>2</sub>MoO<sub>4</sub>, and Li<sub>2</sub>SiO<sub>3</sub> (Shinno, 1987). Synthetic zircons with well-developed plane parallel to the crystallographic c-axis were then handpicked and mounted in Teflon sheet. After polishing with diamond paste and colloidal silica, the zircon crystals were irradiated with <sup>241</sup>Am source for 10 hours at the Institute for Integrated Radiation and Nuclear Science, Kyoto University. Based on the experiments with muscovite, the expected areal ART density is around 0.02 ART/μm<sup>2</sup> (Nakashima et al., 2024)). The irradiated zircon, together with zircon without irradiation, were etched stepwisely with KOH:NaOH=1:1 eutectic etchant at 230°C, which is the same condition with fission track (FT) reveal in zircon, and surface topography was observed by the atomic force microscopy.

**RESULTS:** We run synthesis experiments with two cooling rates after heating at 1270°C for two hours: 10°C/hr and 5°C/hr. The slow cooling experiment resulted in larger crystals and more number of skeletal crystals. AFM observation shows no significant differences between irradiated and non-irradiated samples up to 6 hours etching. This may imply that the etching condition necessary to make ART visible is different from that for FT observation, or that the registration factor for zircon is different from that of muscovite and we need more irradiation to leave a sufficient number of ART on the mineral surface.

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## Concentration Distribution of the Elements in Aerosol Coarse Particle during **Kosa Event**

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The particles which are suspended in the atmosphere, called atmospheric aerosols, have particle size ranging from several tens of nanometers to 100 µm, and the composition differs within each particle size range. Here, we will discuss the elemental composition of soil particles, which are mainly coarse particles (particle size > 2 μm). The soil particles increases its concentration mainly at the time when yellow sand (Kosa particle) arrives. Kosa particles originate in the desert regions of mainland China and arrive in Japan between March and May. From April 16th to 23rd, 2024, Kosa particles also flew into the Kansai region, and we were able to collect Kosa particles. Even when Kosa particles are not arriving, the area is affected by soil particles originating from China and soil particles around the observation point[1].

Here, we will discuss how the concentrations of Al and Ca (main element of soil particle) change and the changes in particle size distribution when Kosa particles arrive. The atmospheric aerosol particles observed on Kosa arriving (April 16th to 23rd, 2024) were collected at Osaka Metropolitan University (Sakai City, Osaka Prefecture) in one-week period using an Andersen sampler, collecting with the particle size ranges divided into nine ranges (<0.43, 0.43-0.65, 0.65- $1.1, 1.1-2.1, 2.1-3.3, 3.3-4.7, 4.7-7.0, 7.0-11, >11 \mu m$ ) and collected onto polyethylene sheets. The average of coarse particle (PM-C) concentration from April 16 to April 23 was 60 µg/m<sup>3</sup>, about five times the average concentration for other non-Kosa periods (14 µg/m<sup>3</sup>). The elemental composition of the collected atmospheric aerosol was analyzed by neutron activation analysis in neutron

irradiation with Kyoto University Research Reactor. The average concentrations of Al and Ca during the Kosa event (April 16-23) were 3700ng/m<sup>3</sup> and 1700ng/m<sup>3</sup>, respectively, which were 4.7 times (Al) and 3.2 times (Ca) higher than the average concentrations during other periods, which were 790ng/m³ (Al) and 530ng/m³ (Ca). The increase in Ca concentration during the arrival of Kosa particles was not as large as that of PM-C and Al, and it is estimated that the Ca concentration in Kosa particles is lower than that in soil particles near the observation point.

Figure 1 shows the particle size distribution of PM-C, Al, and Ca concentrations during the arrival of Kosa particles. The average particle size and standard deviation obtained

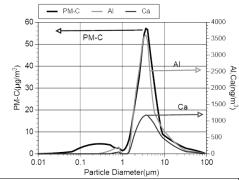


Fig.1 Concentration distribution on particle size for PM-C, Al,Ca

from this distribution are shown in Table 1. When Kosa particles arrive, the average particle size Table 1 Mean diameter and standard deviation obtained from concentration distribution on particle size

and its standard deviation decrease, and it can be assumed that particles with a narrower particle size range are arriving.

	Mean diameter(μm)		Standard deviation of diameter(µm)	
Items	4/16-23	Other periods	4/16-23	Other periods
PM-C	6.5	8.9	5.1	7.4
Al	5.8	8.7	4.4	6.3
Ca	7.0	8.9	5.6	7.5

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# Basic Research on Trace Elemental Analysis of Airborne Particulate Matters Derivated from Concrete in an Environment by INAA & IBA

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**INTRODUCTION:** To clarify the behavior of cesium compound aerosols in sodium-cooled fast reactor (SFR) during severe accident (SA), the aerosol behavior is divided into elementary processes, and simulation experiments will be performed for each process [1] (see Fig.1). In this work, the deposition of CsOH (one of the chemical forms of Cs during SA) to concrete material (one of the SFR structural materials, containing Ca) is focused on. That is, coexistence of these elements containing in the reaction products is investigated with INAA.

**EXPERIMENTS:** The concrete specimen ( $15\text{mm} \times 15\text{mm} \times 30\text{mm}$ ) with 1g of CsOH at its top was placed in the center of a quartz tube ( $\phi$  30 mm). The specimen was heated in an cylindrical electric furnace (Asahi Rika Seisakusho, ARFH-30KC) for 30 minutes at a temperature of 800 °C under atmosphere of relative humidities: 0% and 80% in Ar. After heating, the top surface (reaction products) of specimens were grinded with abrasive paper (#60) to obtain fine powder for INAA (30 s irradiation at PN6 of KUR). These powder specimens (amount for 1mm thickness) were placed in polyvinyl bags, and were sealed.

RESULTS&DISCUSSION: During irradiation, neutron absorption reactions will be happened to make gamma emitter species as shown as Cs-133+n→Cs-134 and Ca-48+n→Ca-49. Radioactive species of Cs-134 and Ca-49 have half-lives of 2.06 y and 8.7 min, respectively. It is interest in the coexistence behavior of Cs and Ca, therefore gamma rays emitted from Cs-134 and Ca-49 were measured by Ge semiconductor detector. Generally, when Ca is present in rock materials such as sand or aggregate of concrete materials, it is incorporated into the silt structure that they possess. However, when Cs enters this location, it is believed that Ca gives up its space and is replaced by Cs [2]. Fig.2 shows a spectrum measured for a concrete specimen at full range 4 MeV instead of normal full range of 2 MeV. The depth profile for Ca-49 for a specimen heated at 800 °C for both humidity are identical, in which those intensities were high near the surface and those were low at at deep positions. This trend is same as in the Cs penetration profiles in concrete materials. If site-exchange reaction happens between free Cs, and Ca located at the silt structure, the depth profiles would show opposite trend each other. This behavior will be continuously researched by obtaining more precise measurements with INAA and/or other analysis methods.

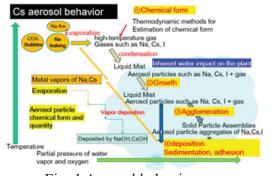


Fig. 1 Aerosol behavior map.

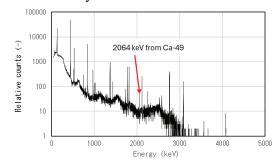


Fig. 2 A spectrum from an irradiated concrete

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## Absorption of alkali metal ions by white radish sprouts (VI

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**INTRODUCTION:** Radioactive <sup>137</sup>Cs, which was released in the Fukushima Daiichi Nuclear Power Plant accident, still remains and is the cause of reputational damage. It is considered necessary to extract <sup>137</sup>Cs from soil in some way. However, <sup>137</sup>Cs has the property that it is strongly adsorbed when it enters soil and cannot be easily extracted. Previous research in our laboratory has shown that when stable isotope cesium is added to contaminated soil, <sup>137</sup>Cs is released from the soil by exchange with <sup>137</sup>Cs adsorbed in the soil[1, 2]. Therefore, in this study, we focused on ion exchange with alkali metal ions as a decontamination method. We created simulated contaminated soil with <sup>137</sup>Cs and performed decontamination using a decontamination solution containing stable isotope alkali metal ions (K<sup>+</sup>, Rb<sup>+</sup>, Cs<sup>+</sup>) with similar properties, to examine which alkali metal ions are effective for decontamination. We also compared conditions for extracting more <sup>137</sup>Cs, such as the addition of nitric acid and dispersants, and temperature changes. In addition, from the perspective of practical application, radish sprouts were grown in the presence of alkali metal ions and the effects of decontamination on plant growth were considered in terms of changes in trace element concentrations.

**EXPERIMENTS:** After radish sprouts were germinated on absorbent cotton moistened with pure water, 10 mL each of CsNO<sub>3</sub> or RbNO<sub>3</sub> solutions of various concentrations and culture medium were added to the absorbent cotton, and the radish sprouts were hydroponically grown and harvested. The trace elements contained in the harvested samples were quantified and compared by neutron activation analysis. Radish sprouts were also grown in soil using androsol and vermiculite to which alkali metal ions had been added, and they were harvested. Neutron activation analysis was then performed to examine the effect of the addition of alkali metal ions on the absorption of various trace elements.

**RESULTS:** In this experiment, radish sprouts were grown hydroponically or in soil in the presence of alkali metal ions, and the effects of decontamination on plant growth were examined. The indicator was the change in trace elements contained in the radish sprouts. When alkali metal ions were added, a decrease in the K content in the radish sprouts was confirmed. This is thought to be because competition with alkali metal ions inhibited the absorption of K into the radish sprouts. On the other hand, the Mn content in the radish sprouts increased with the addition of alkali metal ions. Since Mn is an essential element for plants, if its absorption is promoted, it can be said to have a positive effect on plants. In addition, the increase in Mn content was reduced by washing with pure water, which is thought to allow the increase to be controlled, and it is speculated that the risk of Mn excess can be managed. As changes and trends were observed in the contents of several elements, it is thought that the addition of alkali metal ions (decontamination solution) has an effect on plants.

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## Development of <sup>40</sup>Ar-<sup>39</sup>Ar Dating Equipment in IIRNS, Kyoto University

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INTRODUCTION: Radiometric dating is a valuable tool for unveiling terrestrial and planetary material formation and evolution. The K-Ar and <sup>40</sup>Ar-<sup>39</sup>Ar methods are invaluable in determining the timing of volcanic eruption, cooling process estimation for plutonic rock and resolving the timing of heating events on planetesimals and asteroids (e.g. Fernandes, 2024 [1]). The <sup>40</sup>Ar-<sup>39</sup>Ar dating method and the method with laser heating technique are suitable for tiny samples (e.g. Kelley, 1995 [2] and Hyodo, 2008 [3]). To obtain <sup>40</sup>Ar-<sup>39</sup>Ar dates of the rock and mineral separate samples from terrestrial and extraterrestrial material using the facility in IIRNS, we have continued developing a <sup>40</sup>Ar-<sup>39</sup>Ar dating system, which includes laser-heating gas extraction and gas purification lines. The system was initially designed by Dr. R. Okazaki (Kyushu University) and has been developed for several years. Previous progress reports have described details and the schematic diagram of the system (Nos. 31126, R2079, R3121, R4068, R5122).

**EXPERIMENTS:** In FY2024, we continued assembling the gas extraction and purification system. We added an activated charcoal trap to the gas purification lines for the purpose of transferring sample gas from the sample holder to the gas analyser. We also checked the current condition of the Nd-YAG laser.

**RESULTS:** The activated charcoal traps, which include 0.12 g of activated charcoal grains, were installed in the gas extraction and purification system. When this trap is chilled with liquid nitrogen externally, sample gases are adsorbed on the surface of activated charcoal, and the gases are released when the trap is heated up by hot water. Combining this with valve manipulation can transfer the sample gas within the vacuum line. A Nd-YAG laser (LEE Laser model 812TQ), which was installed in 2019 at IIRNS, had a continuous wave power output of 57 W at the time of installation. However, no laser power output was observed in the latest operation test in March 2025. We must consider replacing the laser equipment to achieve argon isotope measurements in the IIRNS.

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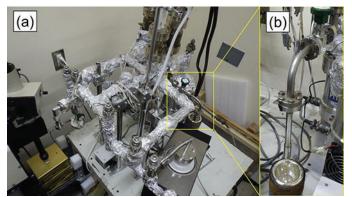


Fig. 1. (a) A photo of an upper view of the gas extraction and purification system. (b) A close-up photo of an activated charcoal trap.

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## Measurement of tritium concentration in modern commercial heavy water reagents used for estimating the migration of radioactive nuclides

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#### **INTRODUCTION:**

Vast forest was markedly contaminated by radioactive plums containing radiocesium in the wide range of the eastern part of Japan in 2011 [1–7]. As forests have an important role in preventing landslides and maintaining the ecological and hydrological system, the destructive forest should be avoided and an appropriately managed tree-felling should be conducted. In our previous field study [1], most of the radiocesium in the tree rings was directly absorbed by the atmospheric direct uptake via the bark and leaves rather than by roots.-We directly measured root distribution of Cryptomeria japonica [3], indicating that mature it is not effective to absorb radiocesium by the root uptake due to the distribution of the fine root [3, 5]. The radiocesium is higher in Konara than in Cryptomeria japonica [1]. The velocity of migration of the nuclide in the unsaturated layer is key to evaluating the absorption of nuclides from the roots. The infiltration rate of soil water in the unsaturated zone is an important factor in evaluating the migration of the nuclides, and heavy water is used to estimate the infiltration rate. However, heavy water contains tritium. We clarified the tritium concentration of commercially available heavy water reagents and examined the environmental impact in field tests.

#### **EXPERIMENTS:**

We selected eleven commercially available heavy water reagents and one <sup>18</sup>O-water (H<sub>2</sub><sup>18</sup>O) deuterium reagent for six markers and one H<sub>2</sub><sup>18</sup>O reagent for one marker available in 2016. These samples will contain high concentration of tritium, a range which fell within direct counting detection limits of modern LSC and does not require any <sup>3</sup>H pre-enrichment. The reagents were distilled 20-fold with ultrapure water. The radioactivity of tritium in the reagents was measured directly by a liquid scintillation counter (LSC) after the addition of a cocktail of liquid scintillation [8].

#### **RESULTS:**

Commercially available the reagents contain tritium ranging from 61 Bq/L to 2.5 x10<sup>3</sup> Bq/L. The concentration in the reagent was higher than that detected in the unsaturated layer in Fukushima [9]. However, the tritium concentration in the reagents was below WHO regulation. After the reagent is used as a field tracer in the environment, since the tritium in the reagents in the surface layer is further diluted by water in the unsaturated zone and then recover to background level quickly.

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## **Determination of Abundance of Rare Metal Elements in Seafloor Hydrothermal Ore** Deposits by INAA Techniques-11: Determination of Barium in Sulfide-sulfate Ore

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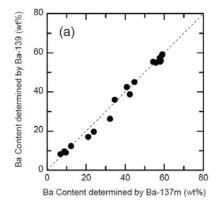
**INTRODUCTION:** We have studied application of INAA technique to quick search for elemental abundance in mineralized samples collected from seafloor hydrothermal deposits. Barite (BaSO4 as chemical formula) is one of important gangue minerals in such deposits, and Ba content of sulfide-sulfate ores widely varies from a few hundred ppm to more than 50 wt%. On the other hand, it is not easy to determine Ba content in ore samples using conventional ICP-MS analysis. In this study, we report determination of Ba content of sulfide-sulfate ore using INAA technique.

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EXPERIMENTS: For determination by the peaks at 165.85 keV of Ba-139 (t½=1.38 hours) and at 661.66 keV of Ba-137m (t½=2.552 minutes), powdered samples of 10-20 mg were irradiated at Pn-3 (thermal neutron flux =  $4.68 \times 10^{12}$  n/cm<sup>2</sup>/sec) for 30 seconds and the gamma ray activity was measured for 3 minutes after adequate cooling time (2-5 minutes). For determination by the peak at 496.26 keV of Ba-131 (t½=11.8 days), powdered samples of 10-20 mg were irradiated at Pn-2 (thermal neutron flux =  $5.50 \times 10^{12}$  n/cm<sup>2</sup>/sec) for 20 minutes and the gamma ray activity was measured for 15 minutes after adequate cooling time (~30 hours). Barium content of samples was calculated based on relative comparison of peak areas with a pure barite crystal grain collected from the Kosaka mine.

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**RESULTS:** Because of low background compared with those of Ba-139 and Ba-131, Ba-137m was revealed as useful for determination of Ba in sulfide-sulfate ore in spite of its rather short half-time. Figure 1 shows comparison of results of quantitative determination using these three nuclides. Barium content determined by Ba-139 sometimes showed lower values than that determined by Ba-137m (Fig. 1a), which would reflect high background of the peak in the low energy range. Barium content determined by Ba-131 sometimes showed about 10-20 % higher values than that determined by Ba-137m (Fig. 1b), which would reflect high background caused by As-76 included in coexist sulfide minerals.



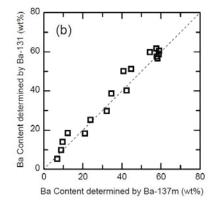


Fig. 1. Comparison of Ba content determined by Ba-131, Ba-137m and Ba-139. (Note that Ba content of pure barite crystal is 58.8 wt%).

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# Experimental evaluation of shock-induced reactions of carbonate minerals: Implications for understanding environmental change by planetary impacts

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#### **INTRODUCTION:**

Planetary impacts by asteroids and comets can dramatically alter the state of the planetary surface due to their high energy density. Carbonate minerals are the main reservoirs of carbon species (e.g., CO<sub>2</sub> and C) on the Earth's surface, and have significant effects on the carbon cycle and atmospheric circulation in the Earth. To model planetary impacts and evaluate impact-induced environmental changes, we need to understand the shock behavior of carbonate minerals under extreme conditions induced by the impact process. In this study, we aimed to evaluate the shock-induced behavior of calcite (CaCO<sub>3</sub>), a potential source of CO and/or CO<sub>2</sub>. To this end, we experimentally investigated the time evolution of calcite during shock compression and decompression processes using an ultrafast time-resolved X-ray diffraction (XRD) coupled with a laser-driven shock compression system.

#### **EXPERIMENTS:**

We conducted the high-power laser shock compression experiments coupled with femtosecond XRD to analyze the structural changes of calcite under high-temperature and high-pressure conditions. Laser-shock XRD experiments were performed in Experimental Hatch 5 at Beamline 3 of the SPring-8 Angstrom Compact Free-Electron Laser (SACLA) facility. The natural calcite crystals used (~1 cm pieces; density, 2.71 g/cm³) were polished to a thickness of  $50 \pm 1~\mu m$  (~4 mm square plates) and were similar to those used by Umeda et al. (2022) for Hugoniot measurements [1]. We investigated the structural evolution of calcite under shock compression up to  $234 \pm 19$  GPa and during decompression from this peak pressure to ambient pressure conditions. The target was composed of 50- $\mu m$  thick polypropylene ( $C_3H_6$ )<sub>n</sub> and a 50- $\mu m$  thick single crystal of calcite (CaCO<sub>3</sub>). An optical drive laser pulse with a rectangular shape (5-ns width) and a wavelength of 532~nm was focused onto the polypropylene, which served as an ablator to generate a near-planer shock wave propagating through the calcite crystal. The X-ray free-electron laser (XFEL) probe pulse with a photon energy of 10.0~keV was focused on the target to a spot size of  $40 \times 13~\mu m$ . The XFEL beam was pointed at the calcite ( $25~\mu m$  center of depth). The X-ray diffraction (XRD) detector had a  $2\theta$  range of  $\sim 20^{\circ}$  to  $\sim 70^{\circ}$ . To estimate the shock velocity and the shock pressure of calcite, velocity interferometer system for any reflector (VISAR) measurements were used to determine the breakout time when the shock wave reached the rear surface of the calcite sample.

#### **RESULTS:**

In this study, we analyzed the structural evolution of calcite during shock compression and decompression as a function of time. The XRD results obtained for the nanosecond shocked calcite provided the direct evidence for the transformation of calcite to the amorphous state during the compression stage at shock pressures above  $86 \pm 7$  GPa and during the decompression stage at shock pressure of  $33 \pm 3$  GPa. Additionally, the results obtained for shock-decompressed calcite indicated that its decomposition reaction (CaCO<sub>3</sub> = CaO + CO<sub>2</sub>) does not occur within the nanosecond shock process [2, 3]. These observations suggests that the shock duration as well as shock pressure and temperature are critical factors that determine shock-induced structural changes [3]. In future, we will conduct shock recovery experiments to investigate the shock-induced deformation structure in post-shock processes.

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## NAA and Ar-Ar dating for small-sized meteorite samples

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**INTRODUCTION:** Meteorites experienced complicated processed prior to the recovery on the Earth, including thermal metamorphism, aqueous alteration, impacts alteration, and irradiation of energetic particles in space. Timing and physicochemical conditions of these events could be recorded in minor element abundances and Ar-Ar ages of meteorites. We have developed and applied the combination analytical method that enables us to carry out gamma-ray measurements and Ar-Ar dating for the same individual samples of sub-milligram mass (e.g., Projects #R3007, R3038, R3127, and R5006). Through the previous projects, we have measured ordinary chondrites and eucrites (Vesta-derived meteorites). In this project of R6083, we applied this analytical method to Martian meteorites.

**EXPERIMENTS:** Samples were plagioclase and maskelynite (shock-melt glass) separates prepared from the A12325 Martian meteorite. Grains of orthoclase [1], wollastonite, and sanidine were used as the standard samples. The meteorite samples and our standard samples were placed in a conical dimple ( $\varphi$ 1, depth  $\sim$  0.5 mm) of a sapphire disk ( $\varphi$ 5.5, 1.5 mm thick), and covered with a sapphire disk ( $\varphi$ 5.5, 0.3 mm thick). Each of the sapphire container was wrapped with pure aluminum foil. These Al-wrapped containers were stacked and sealed in the capsules for the Hydro irradiation. The sample irradiation was conducted in October 2024 under our nominal condition (1MW  $\times$  47 hours + 5MW  $\times$  6 hours). The irradiated samples were recovered and transported to Kyushu Univ. in November after the decays of short-lived nuclides.

**RESULTS:** Our original plan was to conduct two irradiation experiments for NAA and Ar-Ar dating. However, due to the failure of the mass spectrometer for noble gas isotopes, the analysis scheduled for November could not be performed. In addition, the irradiation planned in early 2025 had to be cancelled. The irradiated samples have been preserved, and we plan to conduct noble gas isotope analysis as soon as possible after the repair of the mass spectrometer is completed.

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# Volcanic and Tectonic History of Philippine Sea Plate (South of Japan Revealed by $^{40}$ Ar/ $^{39}$ Ar Dating Technique

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**INTRODUCTION:** The origin and history of spreading of the West Philippine Basin and its tectonic relation to the adjacent Izu-Bonin-Mariana arc in the Philippine Sea Plate remains poorly-understood. Especially, understanding of the temporal and tectonic relationship between the birth of the Izu-Bonin arc and the West Philippine Basin is critical for reconstruction of Philippine Sea plate tectonics, which closely links to subduction initiation of the Pacific Plate along its margin.

The YK22-11S cruise aimed to investigate the origin and age of magmatism and structure of intersection between the spreading center of the West Philippine Basin (CBF Rift) and the Kyushu-Palau Ridge, i.e., remnant Izu-Bonin-Mariana arc. Two Shinkai6500 submersible dives were conducted in the CBF Rift spreading center. One dive (6K1646) observed an ESE-WNW trending ridge, which might be one of the youngest volcanic features in the rift. This dive recovered sam-ples of basaltic lava flows, quite often showing morphological characteristics of pillow lava. An-other dive (6K1647) in the rift examined a section of the CBF Rise, further west from the inter-section point than the previous dive. This dive again recovered a number of basalt samples of pil-low lava origin and also hyaloclastite. This year we determined <sup>40</sup>Ar/<sup>39</sup>Ar ages for 20 basaltic rock samples which were recovered during the two dives mentioned above.

**EXPERIMENTS:** Ages of the igneous rocks were determined using the <sup>40</sup>Ar/<sup>39</sup>Ar dating facility at the Geological Survey of Japan/AIST. 10-15 mg of phenocryst-free groundmass, crushed and sieved to 180 – 250 μm in size, was analyzed using a stepwise heating procedure. The samples were treated in 6N HCl for 30 minutes at 95°C with stirring to remove any alteration products (clays and carbonates) present in interstitial spaces. After this treatment, samples were examined under a microscope. Sample irradiation was done either at the Kyoto University Reactor (KUR). The neutron irradiation was performed for 10 h at the hydro-irradiation port under 1 MW operation, where thermal and fast neutron fluxes are 1.6 x 10<sup>13</sup> and 7.8 x 10<sup>12</sup> n/ cm<sup>2</sup> s, respectively, or for 2 h under 5 MW operation, where thermal and fast neutron fluxes are 8.15 x 10<sup>13</sup> and 3.93 x 10<sup>13</sup> n/cm<sup>2</sup> s respectively. Argon isotopes were measured in a peak-jumping mode on an IsotopX NGX noble gas mass spectrometer fitted with a Hamamatsu Photonics R4146 secondary electron multiplier.

**RESULTS:** 10 samples from the dive 6K1646 show age range between 28.3 and 30.9 Ma. These samples show geochemical variation with age, which shows enrichment in incompatible elements and radiogenic isotopes with time from relatively depleted MORB like composition. These data can be interpreted to show magmatic process at the last stage of spreading or rifting along the spreading center. As the extension along the spreading center was wanning, degree of mantle melting decreased, which causes increase in content of the incompatible elements and also preferential melting of mantle domain enriched in radiogenic isotopes. On the other hand, 10 samples from the dive 6K1647 show age range between 31.5 and 33.1 Ma, clearly older than those from 6K1646. Source of these basalts are estimated to have more enriched mantle, with similarity to EM-2 mantle component. These temporal geochemical variations discovered along the CBF Rift spreading center can be used to decipher processes when the seafloor spreading center approaches and clashes with volcanic arc.

## Amorphization of NaAlSi<sub>3</sub>O<sub>8</sub> feldspar by shock compression

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**INTRODUCTION:** Strongly-shocked stony meteorites record hypervelocity collision events on their parent asteroids. Such meteorites often contain NaAlSi<sub>3</sub>O<sub>8</sub>-rich feldspar (albite) as a major mineral constituent. Based on X-ray diffraction and infrared spectroscopy of samples from laboratory shock experiments, it has been shown that albite begins to metastably transform into dense glass at ~34 GPa and completes at ~45 GPa [1-2]. This process is one of the indicators of the shock pressure, but the microstructural changes of albite during amorphization and the structure of the dense glass have not been sufficiently investigated. For a detailed understanding of the pressure-induced amorphization, we demonstrate shock recovery experiments of albite and X-ray diffraction (XRD) and transmission electron microscopy (TEM) of the recovered samples.

**EXPERIMENTS:** Single crystals of albite were processed into thin sections with smaller than 10 mm in size and with 1.0 mm in thickness. Each section was placed into a stainless steel (SUS304) container, and shock loaded using a single-stage propellant gun at 12 GPa (Shot1) and 29 GPa (Shot2). In one of the shots, the space between the inner wall of the container along the compression axis and the sample was filled with Al powder, to obtain a higher cooling rate than the shots without Al power, shocked at 29 GPa (Shot3). The central portions of the recovered thin section samples were first observed by TEM operated at 200 kV accelerating voltage, and then analyzed by XRD (Rigaku Ultima IV) at 1.6 kW X-ray power.

**RESULTS:** Under TEM, the Shot1 sample does not show any deformation and transformational features. Meanwhile, the Shot2 sample is mostly amorphized. Relict plagioclase crystal appears as

thin lamellae less than 1  $\mu m$  thick. Electron diffraction patterns show that the lamellae are oriented on the (010) plane of albite. The Shot3 sample shows the same microstructure as Shot2. These results suggest that the onset pressure of amorphization of albite is lower than 34 GPa which value was previously es-

timated by bulk sample analysis, regardless of the cooling rate after shock heating.

Shot1 and 2 were also analyzed by XRD. Both of the samples show broad First Sharp Diffraction Peak (FSDP) at ~0.2 (Å-1). The FSDP of Shot #2 is 0.006 (Å-1) greater than that of Shot1, indicating a denser glass structure. The results suggest that the FSDP shift is a potential indicator of shock pressure.

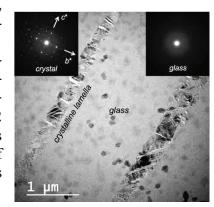


Fig. 1. TEM image of albite shocked at 229 GPa.

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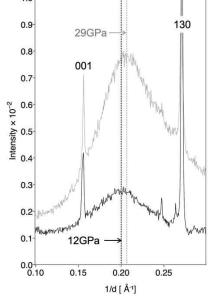


Fig. 2. XRD patterns of albite shocked at 12 and 29 GPa. Sharp diffraction peaks are due to weakly shocked part of albite at the edges of the sample.

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### Evaluation of Cs and Sr dynamics in forest ecosystems

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**INTRODUCTION:** Forests contaminated by radionuclides released in the Fukushima Daiichi nuclear accident have yet to be decontaminated except for an area about 20 meters from the forest edge and there are areas of high air dose rates scattered throughout the forest. Because forests make up about 70% of Fukushima Prefecture, decontamination of forests more than 20 meters from the forest edge is necessary to reduce the air dose rate in the forests and restore life and industry surrounding the forests. But it is impossible to decontaminate the entire forest. In this study, in order to consider the feasible scope and methods of decontamination according to local conditions, we aim to develop a model that can quantitatively evaluate behavior of radioactive Cs and Sr in forest ecosystem. This year, we measured Cs in forest slope soil, analyzed the movement of Cs in the slope soil through experiments. The results of the measurement and experiment were reflected in the parameter values of our previous developed model for the transport of radioactive Cs in the forest slope and air dose rate estimation model.

**EXPERIMENTS:** We measured the distribution of Cs concentration in forest slope soil collected from Mt. Mari in Kawauchi Village, Fukushima Prefecture on Nov. 1, 2024. The results show that the concentration does not simply increase the further down the slope, is the highest in the 0-5 cm

layer and the second highest in the 5-10 cm layer.

A Soil column was constructed using the above soils. The equipment is shown in Fig.1. The simulated rainfall was applied to it and measure the runoff rate of Cs on the soil slope.

Using the measurement data obtained in the experiment, Cs runoff rate in the surface slop soil was calculated to be  $6.93 \times 10^{-9}$  s<sup>-1</sup>.

RESULTS: The results of the measurement and experiment were reflected in the parameter values of our previous developed model for the transport of radioactive Cs in the forest slope and air dose rate estimation model. As a result, we could roughly reproduce the measured Cs concentration in the slope soils by model calculation. Using the calculated Cs concentration in soil, future air dose rate in the monitored area were also simulated using our previous air dose rate estimation model.

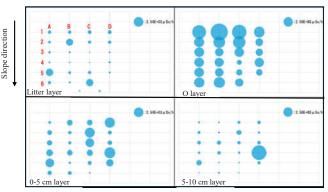


Fig. 1. Measurement results of Cs concentration in soil

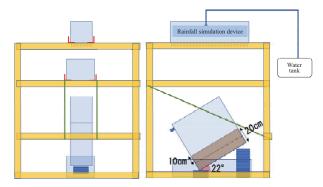


Fig. 2. Experimental equipment

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#### CO5-13

# Fission Track Analysis of Fissile Material in Fallout Sample Collected on Daigo Fukuryu Maru

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**INTRODUCTION:** In the event of nuclear weapon explosions or nuclear disasters, particles originating from nuclear fuels are formed under conditions of high temperature and high pressure. During the Fukushima Daiichi Nuclear Power Plant accident, particles containing high concentrations of radioactive Cs have been discovered in environmental samples [1, 2]. Analysis of these particles provides important information for estimating the conditions inside the reactors. We have developed a method for detecting particles containing high concentrations of fissile substances such as <sup>235</sup>U and <sup>239</sup>Pu in environmental samples. This detection method combines fission track analysis using a research reactor with heavy liquid separation [3]. In the previous work, we analyzed soil samples presumed to contain fallout materials from the Hiroshima atomic bomb using this method. It was found that fission track analysis with appropriate neutron irradiation conditions could clearly distinguish particles originating from the A-bomb from natural uranium in the environment [4]. However, these particles could not be identified and analyzed using a SEM because their size was smaller than the spatial resolution of the SEM used in that study. This study aims to detect fallout particles containing fissile material, focusing on the detection of larger particles, by analyzing radioactive fallout samples from the Bravo nuclear weapon test.

**EXPERIMENTS:** The fallout sample, mixed with rust, was collected from a drum can placed on the Daigo Fukuryu Maru. After the sample was spread on the medicine wrapping paper, distribution of the radioactive isotopes was observed by autoradiography. Most of the sample was then collected in a sample container. Small particles remaining on the surface of the paper were subsequently collected on a paraffin film by pressing the film against it. The fission track detector was wrapped in the paraffin film so that the collected particles were in contact with the detector, and then irradiated with approximately  $3 \times 10^{12} \text{ n/cm}^2$  of thermal neutrons using the pneumatic irradiation facility (TC-Pn) at Kyoto University Research Reactor. After irradiation, the track detector was etched, and the resulting tracks were observed using an optical microscope.

RESULTS: The image of the radioactive material distribution obtained by autoradiography indicated that the radioactive fallout in the sample was not homogeneous, as there was no correlation between particle size and radioactivity. Observations of the detector surface revealed multiple clusters of tracks. Similar analysis of soil samples collected in Hiroshima also detected clusters of tracks. These clusters are thought to have been produced by fine particles containing high concentrations of enriched uranium derived from the atomic bomb. In this study, the number of tracks in each cluster ranged from tens to hundreds. Furthermore, the ratio of the number of tracks to the irradiated neutron fluence was similar to those observed for Hiroshima soil samples. This indicates that the radioactive fallout collected on the Daigo Fukuryu Maru contains fissile material at a level comparable to that in the enriched uranium particles derived from the atomic bomb. Based on the observed track distribution, the particle size is estimated to be smaller than several µm. Future analysis of the samples using SEM/EDX will be conducted to examine their appearance and elemental composition.

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## Three halogens (Cl, Br, and I) in mantle-derived samples

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**INTRODUCTION:** Among the four halogens, F, Cl, Br, and I are present in the environment as stable elements. Halogens are highly mobile, volatile, and incompatible elements in a range of geochemical processes. Due to their chemical characteristics, these elements have been widely used to trace the water cycle in subduction zones, understand the mechanism of magma degassing, and evaluate the recycling of solid earth materials in the crust and mantle. Despite the importance of halogens in geochemical processes, the determination of halogens in geological samples is scarce. The reason for this is the difficulty in determining these elements due to their volatility and low concentrations in geological samples. In the previous studies [1,2], USGS and GSJ geochemical reference materials were analyzed by both instrumental neutron activation analysis (INAA) and radiochemical neutron activation analysis (RNAA) to evaluate the accuracy of RNAA data. In this study, we analyzed mantle-derived samples to evaluate the applicability of our RNAA procedure to geological samples.

**EXPERIMENTS:** Two MORB samples were analyzed in this study. Together with these two MORB samples, USGS geochemical reference samples (BCR-2 and DTS-2b) also analyzed as control samples. The analytical procedure of RNAA is described in Ebihara et al. [1]. Samples were weighed into plastic vials and an appropriate amount of each halogen standard solution dropped on a filter paper was sealed in polyethylene as halogen reference samples. Both of these were irradiated for 20 min at the Kyoto University Research Reactor Institute. Know amounts of three halogens and Mn carriers were added into a Ni crucible with the irradiated reference materials and then were fused in NaOH. The fuse cake was digested in water, and a hydroxide precipitate was removed from the supernatant, which contained the halogens. After acidification of the supernatant, iodine was precipitated as PdI<sub>2</sub>, and Cl and Br were precipitated as a mixture of AgCl and AgBr precipitates. These precipitates were subjected to the measurement of <sup>128</sup>I, <sup>38</sup>Cl, and <sup>82</sup>Br. The chemical yields of the three halogens during radiochemical separation were determined using the reactivation method. To correct the effects of neutron flux gradient and neutron attenuation, samples were sandwiched by a reference standard, which was prepared by dropping a known amount of I standard solution on a filter paper.

**RESULTS:** For the geochemical reference materials (BCR-2 and DTS-2b), our RNAA data of the three halogens (Cl, Br, and I) were consistent with those reported in the previous study [1], confirming that our RNAA values in this study are reliable. The two analyzed MORB samples have similar trace element abundances such as REE to each other, while significant differences in the three halogens can be seen. One of the MORB samples has approximately ten times higher abundances of Cl and Br compared with the other. A similar trend cannot be seen in I. In a plot of Br/Cl vs. I/Cl, our two MORB samples are plotted on the trend observed from the previous study [3]. The sample with higher Cl and Br abundances shows a lower I/Cl ratio compared with the other MORB samples. This lower I/Cl ratio indicated that this sample was contributed by seawater.

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# Noble gases and halogens in serpentinite-related metasomatic rocks in the Cretaceous Sanbagawa metamorphic belt, SW Japan

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INTRODUCTION: Since some of the halogens (fluorine, chlorine, bromine, and iodine) are converted to corresponding noble gas isotopes by neutron irradiation and detection limits for noble gas isotopes by mass spectrometry are low (down to ten thousand atoms), they can be analyzed with high sensitivity by using neutron irradiation and noble gas mass spectrometry [1]. Deep earthquakes are generally recognized where subducting oceanic plates are in contact with the shallow mantle. High fluid pressure is commonly invoked as an essential factor in the generation of slow earthquakes, and the associated fluids are thought to be derived from the breakdown of hydrous minerals such as chlorite and serpentine in the subducting slab and the base of the mantle wedge. When considering how such fluids may cause earthquakes, it is crucial to consider both the quantities and fluid paths. Serpentinite developed along the base of the mantle wedge may act as an effective channel for the fluids along the subduction boundary. Numerous serpentinite bodies are exposed throughout the Sanbagawa belt. We analyzed noble gases and halogens in the two separate kilometer-scale serpentinite bodies, the Shiraga Yama (SY) and Kamabuse Yama (KY) bodies, which ascended from depths of ~35 km and ~25 km, respectively [2], to help identify the source of fluids related to hydration and material transport.

**EXPERIMENTS:** The samples of 5-50 mg each and standards for neutron fluence were wrapped with aluminum foil and put in aluminum capsules of  $\phi10$  mm x 30 mm. The capsules were irradiated with neutrons in KUR. After the irradiation, the samples were sent to the University of Tokyo. The samples were loaded into an ultra-high-vacuum, noble gas extraction, purification, and separation line. Noble gases were extracted from the samples by heating up to  $1800^{\circ}$ C, purified with hot Ti-Zr getters, separated into each noble gas with temperature-controlled cold traps, and then their isotope compositions were determined with a noble gas mass spectrometer [1]. The thermal and fast neutron flux was estimated from the production of  $^{39}$ Ar and  $^{38}$ Ar from  $^{39}$ K and  $^{37}$ Cl in the Hb3gr hornblende standard, in which K and Cl contents have been determined.

RESULTS: The results for KY are more complex, with distinct fluids responsible for serpentinization (1) and metasomatism (2). The metasomatism can be further divided into a chlorite-forming stage (2-1) and a later talc-forming stage (2-2). The sources for the fluids involved in each of these stages were likely derived from (1) altered oceanic crust, (2-1) altered oceanic crust + sedimentary porosity, and (2-2) sedimentary porosity + serpentinized slab. Combining the results from SY and KY suggests that channeling of subduction fluids in the Sanbagawa subduction zone was important at depths of around 35 km, which is consistent with the observation of subduction fluids in the Higashi-akaishi peridotite body exhumed from a depths of 100 km [3]. On the other hand, although the fluid channel was less effective at shallower levels, it was possible to rise close to the surface, as preserved in the subduction mélange in the Shimanto accretionary complex [4]. The change in the source region of the fluids with time, as shown in KY, suggests fluid flow may become more channelized as a shear zone develops along the subduction interface.

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## Validation of acid compounds by simulating a model screen (made of red copper leaf)

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**INTRODUCTION:** Analyses by using SEM and XRD of the "Black Rain" traces on a gold folding screen (red copper leaf: copper alloy foil containing zinc), discovered in Hiroshima in 2022, showed that copper sulfate hydrate was presumed to be mixed. However, copper nitrate compounds were undetectable. Therefore, we conducted a simulation experiment of a model screen using sulfuric acid and nitric acid, being contained in the "Black Rain," to verify the results.

**EXPERIMENTS:** After dropping 20  $\mu\ell$  of pH 1 solution containing sulfuric acid, nitric acid, and mixed acids of both in Chinese ink on a model screen piece "RSY-001" (red copper leaf), each dissolved part was observed with an optical microscope and analyzed with a fully automated X-ray diffractometer (Rigaku SmartLab Cu K $\alpha$  source, tube voltage 40 kV, tube current 200 mA, micro-incident optics, scan speed 0.5°/min) in 10 days and 4 months exposed indoors.

**RESULTS:** The black traces were observed under an optical microscope (reflected light/transmitted light images; Fig. 1), and dissolution of the red copper leaf was observed in both light images. Analysis by XRD of the samples after 10 days of the experiment revealed copper sulfate hydrate peaks in the case of sulfuric acid, and copper nitrate compound-derived peaks in the case of nitric acid (Fig. 2). The XRD analysis of the samples exposed for 4 months showed that the peaks decreased in all cases, and in the case of the mixed acid, the copper sulfate hydrate peak remained, but the one from the copper nitrate became minute. The copper nitrate compound could not be detected owing to its deliquescence [1].

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**ACKNOWLEDGMENTS**: We are grateful to the staff of the Hiroshima Peace Memorial Museum, especially the curatorial staff and the donor for permission to cut the gold folding screen. We thank Rekiseisha Co.,

	HNO <sub>3</sub>	H <sub>2</sub> SO <sub>4</sub>	Mixed acid
pH1			
Reflected light image			
Transmitted light image			

Fig. 1. Reflected and transmitted light images of a model screen after dropping acid solutions.

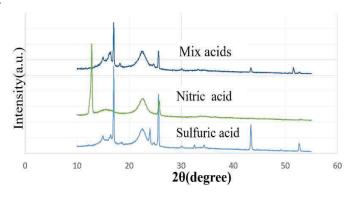


Fig. 2. XRD spectra of products of model screens.

Ltd. for a model screen. Lastly, Prof. Takuo Okuchi, KURNS is acknowledged for his guidance in the XRD analysis.

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