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Editors: T. Moriguchi, Y. Yamato, M. Matsumura, T. Yoshida,
K. Ishimoto, T. Manako and K. Sasa

UTTAC ANNUAL REPORT is a series of issues, which describes annual research activities at Tandem Accelerator Complex, Center for Research in Radiation, Isotopes, and Earth System Sciences (CRiES), University of Tsukuba.

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PREFACE

This annual report presents the research and development (R&D) activities conducted at the University of Tsukuba Tandem Accelerator Complex (UTTAC) during fiscal year 2025 (1 April 2025–31 March 2026). These activities include accelerator-based R&D using the 6 MV tandem accelerator and the 1 MV Tandetron accelerator, as well as radioisotope-based R&D employing positron annihilation spectroscopy and Mössbauer spectroscopy. Results from several industry-led research projects are not included in this report, mainly because of patent-related considerations.

In fiscal year 2025, with support from the KEK IINAS-NX program, we developed metaverse-based educational materials for the 6 MV tandem accelerator using VRChat (Fig. 1). We also participated in the Metaverse Accelerator Museum (MAM) and held “KEK Day: An Invitation to Accelerators—Metaverse Accelerator Museum” at the University of Tsukuba on 19 March 2026 (Fig. 2). Detailed three-dimensional models of the interior of the 6 MV tandem accelerator and its ion source were created for use in facility tours for high school students and practical training for university students. During fiscal year 2025, the facility received a total of 280 visitors. We plan to further develop these educational materials by integrating the VR accelerator simulator with the Metaverse Accelerator Museum.

1 October 2026

Editorial Board

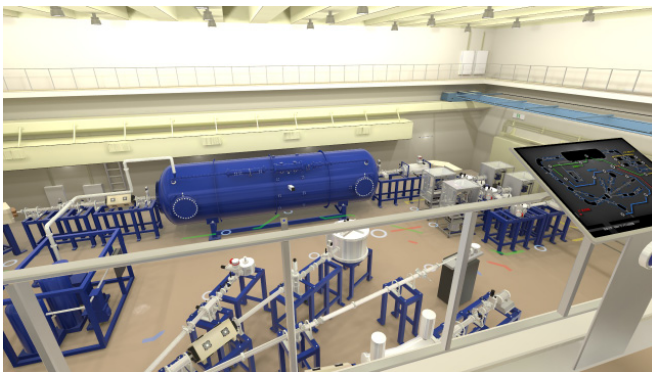


Fig. 1 Metaverse-based educational materials for the 6 MV tandem accelerator using VRChat.



Fig. 2 Poster of “KEK Day: An Invitation to Accelerators—Metaverse Accelerator Museum”.

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1.

ACCELERATOR OPERATION AND RELATED STUDIES

1.1 Accelerator operation 2025

K. Ishimoto, S. Ishii, T. Takahashi, Y. Yamato, T. Yoshida, T. Manako, M. Matsumura, T. Moriguchi, K. Sasa

University of Tsukuba Tandem Accelerator Complex (UTTAC) conducts the maintenance and operation of the tandem accelerator facility consisting of the 6 MV Pelletron tandem accelerator and the 1 MV Tandetron accelerator for collaborative research both inside and outside the University of Tsukuba.

1 MV Tandetron accelerator

The 1 MV Tandetron accelerator has 2 negative ion sources and 4 beamlines. The operating time and the experimental beam time of the 1 MV Tandetron accelerator were 1,725.1 and 362.2 hours, respectively, during the total service time in FY2025. The 1 MV Tandetron accelerator operated for 101 days in FY2025, with 508 participants and 49 experimental runs conducted. Figures 1 and 2 show the classification of accelerated ions and irradiation times by application, respectively. Since the number of experiments with short irradiation times increased in FY2025, the distribution of operating days is also shown in Figure 3. At the end of March 2026, the total number of approved research programs was 6 for on-campus and 3 for off-campus. The instability in terminal voltage, which has been reported since FY2022, was occasionally observed again in FY2025, even though the issue was largely mitigated by allowing sufficient adjustment time before accelerator startup.

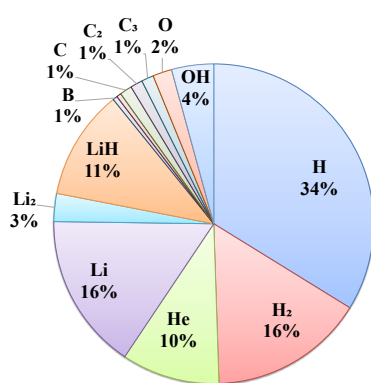


Fig. 1. Accelerated ions by the 1 MV Tandetron accelerator in FY2025.

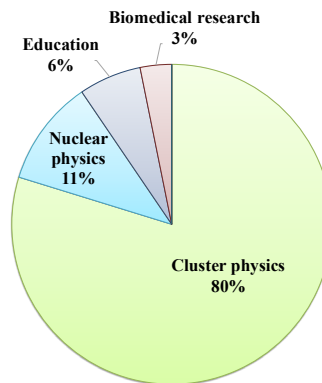


Fig. 2. Irradiation time by the 1 MV Tandetron accelerator in FY2025.

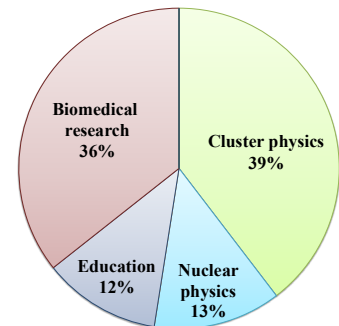


Fig. 3. Operating days of the 1 MV Tandetron accelerator in FY2025.

6 MV Pelletron tandem accelerator

The 6 MV Pelletron tandem accelerator has 5 negative ion sources and 12 beamlines. The operating time and the experimental beam time of the 6 MV Pelletron tandem accelerator were 1,321 and 1,124.9 hours, respectively, during the total service time in FY2025. The 6 MV Pelletron tandem accelerator operated for 137 days in FY2025, with 1,132 participants and 79 experimental runs conducted. Figure 4 shows the beam time histogram with respect to the terminal voltage. Figures 5 and 6 show the classification of the accelerated ions and of the experimental purposes, respectively. At the end of March 2026, the total number of approved research programs was 8 for on-campus and 7 for off-campus. The operating time decreased by

approximately 11% compared to that in the previous fiscal year [1]. This reduction was primarily attributable to the allocation of approximately one month for training new staff members.

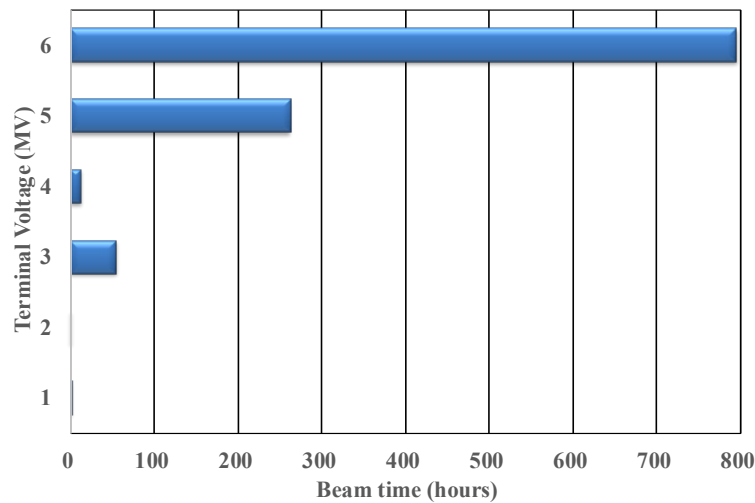


Fig. 4. Beam time histogram as a function of the terminal voltage for the 6 MV Pelletron tandem accelerator in FY2025.

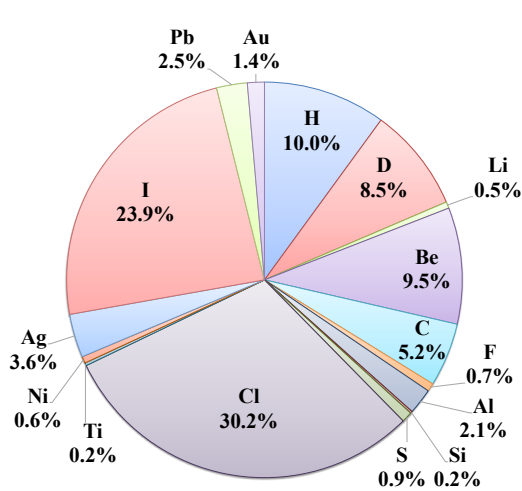


Fig. 5. Accelerated ions by the 6 MV Pelletron tandem accelerator in FY2025.

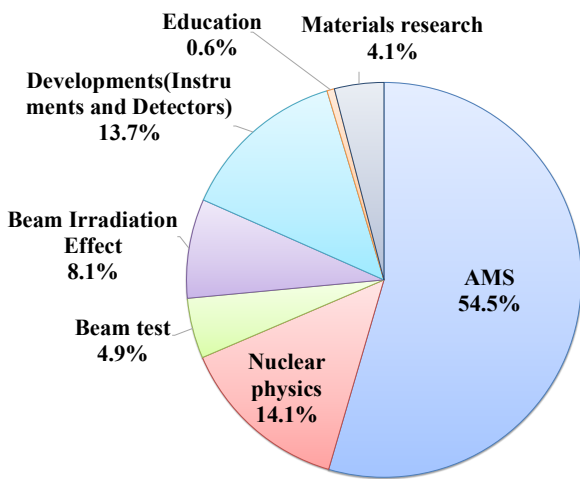


Fig. 6. Experimental purposes of the 6 MV Pelletron tandem accelerator in FY2025.

Reference

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1.2 Development of ion irradiation techniques for space-use device irradiation testing equipment

T. Yoshida, T. Takahashi, Y. Yamato, T. Manako, K. Sasa

Since space environments contain cosmic radiation, prior assessment of single-event effects (SEE) must be performed for space-use devices. In this study, ion irradiation techniques necessary for SEE testing were developed. At the University of Tsukuba Tandem Accelerator Complex (UTTAC), ion beams with linear energy transfer (LET) values on the order of several tens of MeV/(mg/cm²) can be provided. Beam tests using ions such as Au and Si have been conducted [1]. The facility has also been continuously utilized by external organizations, including JAXA [2].

Previous experimental results have shown that faint beams of 72 MeV Si (beams attenuated to an extremely low intensity of several hundred particles per second) exhibit multiple peaks in the energy spectrum measured by a solid-state detector (SSD) [3]. Such multi-peak beams are not suitable for irradiation tests. Therefore, experiments were conducted to determine the conditions required to obtain a single-peak beam. In the present work, we focused on the charge-exchange method. Table 1 lists faint beams that exhibit a single peak. Open circles (○) indicate beams produced by gas charge exchange, while filled circles (●) indicate those produced by foil charge exchange. Ion species with lower charge states than those indicated are also available. These results indicate that a single-peak beam can be obtained depending on the combination of ion species and charge-exchange method. Further investigations are planned to determine the optimal conditions for other ion species.

Table 1. Faint beams that exhibit a single peak under different charge-exchange conditions.

Charge state	Energy (MeV)	H	Li	C	O	F	Al	Si	S	Ag	Au
14	90									○	○
13	84										
12	78										
11	72								●		
10	66							●			
9	60						○				
8	54				○	●					
7	48										
6	42			○							
5	36										
4	30										
3	24		○								
2	18										
1	12	●									

○:gas charge exchange ●:foil charge exchange

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- [1] M. Sataka et al., UTTAC Annual Report 2023, UTTAC-93 (2024) 5.
- [2] M. Takahashi et al., UTTAC Annual Report 2024, UTTAC-94 (2025) 50.
- [3] M. Sataka et al., UTTAC Annual Report 2024, UTTAC-94 (2025) 3.

2.

NUCLEAR AND COLLISION-BASED RESEARCH

2.1 Precise magnetic moment measurements of unstable nuclei ^{25}Al and ^{29}P

A. Ozawa, R. Iwamoto, H. Kobayashi, M. Mikawa, M. Mitsui, T. Moriguchi, Y. Saito, I. Thompson, Y. Yamato, A. Yano

The nuclear magnetic moment (μ) is significantly sensitive to nuclear structure. Since the nuclear structure information for unstable nuclei is very limited, measurement of μ for unstable nuclei is indispensable. The μ values of the unstable nuclei ^{25}Al and ^{29}P have already been measured [1]; however, the associated uncertainties can be reduced. In the present study, we aim to precisely determine μ of ^{25}Al and ^{29}P based on μ of ^{20}F , which is known more precisely than those of ^{25}Al and ^{29}P . The experiment was performed using the 6 MV tandem accelerator at UTTAC. We used a Lamb-shift-type polarized ion source to produce polarized ^{20}F , ^{25}Al , and ^{29}P . Measurements of μ were performed by the β -NMR method. In this method, if we measure μ of ^{20}F and ^{29}P in the same static magnetic field, μ of ^{29}P can be obtained by

$$\mu(^{29}\text{P}) = \mu(^{20}\text{F}) \times \frac{\omega(^{29}\text{P})}{\omega(^{20}\text{F})} \times \frac{I(^{29}\text{P})}{I(^{20}\text{F})} \quad (1)$$

where ω is the observed Larmor frequency and I is the nuclear spin of each nuclide. After measurements of μ for ^{29}P , the same procedure was applied to determine μ of ^{25}Al based on μ of ^{29}P . The preliminary results of μ of ^{29}P (^{25}Al) based on μ of ^{20}F (^{29}P) are shown in Figs. 1(a) and 1(b), respectively. In the ^{29}P measurements, we measured μ of ^{20}F using a CaF_2 target and μ of ^{29}P using Si(p-type, low impedance) and Si(n-type, low impedance). In the ^{25}Al measurements, μ of ^{29}P and ^{25}Al were measured using Si(p-type, high impedance) and Si(n-type, high impedance). Note that no correction for systematic shifts has been applied to these preliminary results. Although the present results still deviate from the previous values, they are mutually consistent and the error bars are smaller than those of the previous measurements.

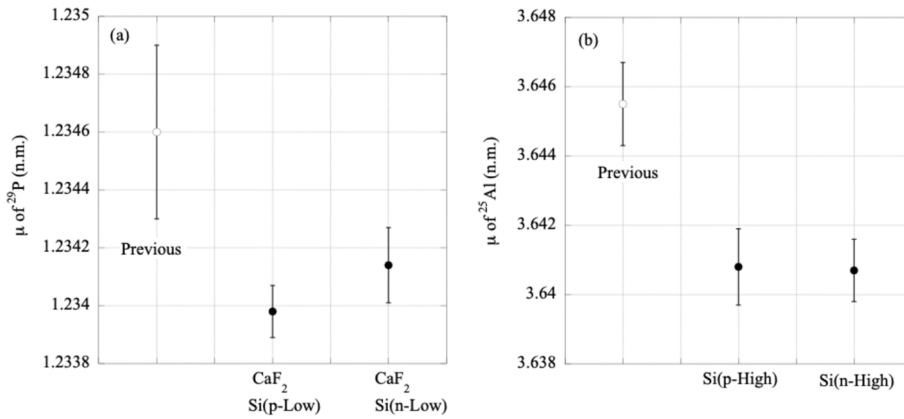


Fig. 1. Preliminary results of μ for (a) ^{29}P and (b) ^{25}Al . Previous values [1] are shown by open circles. Present results are shown by closed circles.

References

[1] N.J.Stone, At. Data Nucl. Data Tables 90, 75 (2005).

2.2 Measurement of a low-energy reaction cross-section of deuteron on a carbon target (II)

K. Watanabe¹, H. Ito¹, Y. Kikuchi¹, D. Nagae¹, S. Nishizawa¹, R. Takano¹, T. Yamaguchi^{1,2}, I. Yasuda¹, W. Yu¹, T. Moriguchi, A. Ozawa, A. Yano, M. Mikawa, M. Mitsui, H. Kobayashi, R. Iwamoto, Y. Saito

This study presents ongoing measurements of the low-energy reaction cross-sections for deuteron–carbon interactions [1]. The experiment extends the methodology of matter-radius determination via reaction cross-sections to the low-energy regime. In 2024, we conducted a one-day experiment at the A7 beamline and identified deuterons and protons by using the ΔE – E method with a silicon detector telescope. In 2025, we continued the experiment with the following updates:

- The deuteron energies were adjusted to be identical for target-in and target-out runs. This was necessary because the reaction events in the detector telescope were canceled using the transmission method. The applied energies for the target-in and target-out runs were 12.00 MeV and 11.02 MeV, respectively; the energies after passing through the reaction target were consistent with each other within the experimental uncertainties.
- Pile-up rejection electronics were introduced to enable the unambiguous identification of deuterons. The fraction of pile-up events decreased from 0.41% to 0.074%.
- The counting loss due to the elastic scattering of deuterons was evaluated by varying the position of the detector telescope. Three configurations were employed with scattering angles of (a) -18° to 18° (on the beam axis), (b) 12° to 42° , and (c) 37° to 55° . Figs. 1(a) and 1(b) correspond to configurations (a) and (b), respectively, showing the energy-loss correlations of the two silicon detectors. In configuration (c), almost no events were recorded. In Fig. 1(b), only a small number of scattered deuterons are observed, and the stripped protons are clearly visible along the hyperbolic locus. Data analyses are currently in progress.

A low-energy component of the deuterons was consistently observed in the first detector of the silicon telescope. This may be attributed to the upstream beam-energy distribution. Further investigations and improved statistical analyses are planned for future experiments.

References

- [1] K. Watanabe et al., UTTAC Annual Report 2024, UTTAC-94 (2025) 5.

¹Department of Physics, Saitama University

²Visiting associate professor appointment

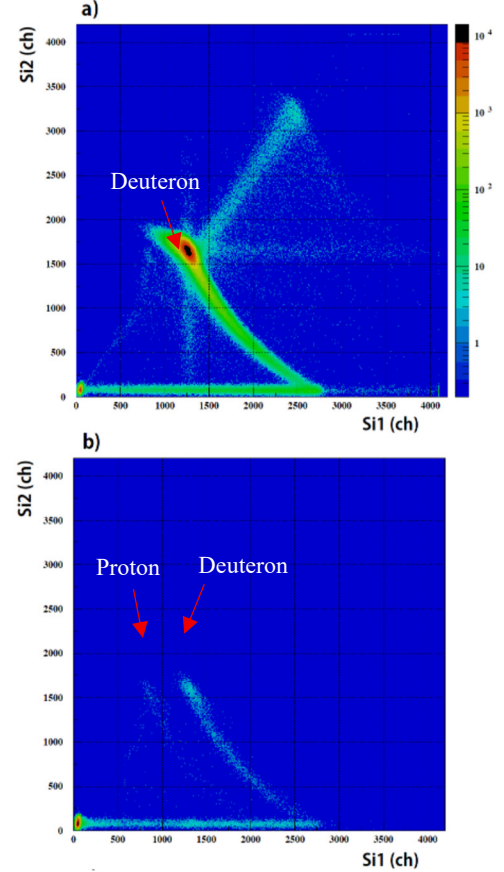


Fig. 1. Two-dimensional plots of the silicon ΔE – E telescope. The scattering angle ranges are (a) from -18° to 18° and (b) from 12° to 42° .

2.3 Extraction efficiency of ^{14}C produced via the $^{11}\text{B}(^4\text{He}, \text{p})$ reaction

T. Moriguchi, M. Mikawa, A. Ozawa, K. Sasa, T. Takahashi, M. Matsumura, T. Yoshida, S. Ishii, K. Ishimoto, T. Manako, A. Yano, M. Mitsui, H. Kobayashi, R. Iwamoto, Y. Saito

We are developing low-energy radioactive nuclear beams of long-lived nuclei [1]. A key advantage of directly extracting long-lived nuclides from an ion source is that beams with a narrow energy distribution can be obtained. In FY2025, we focused on ^{14}C , which exists naturally in trace amounts and is widely measured by accelerator mass spectrometry (AMS), for example in radiocarbon dating. As a research subject using a ^{14}C beam, the $^{14}\text{C}(\text{n}, \gamma)^{15}\text{C}$ reaction is important for neutron-induced CNO cycling in AGB stars and inhomogeneous Big Bang nucleosynthesis, while significant discrepancies still remain among experimental values of the low-energy cross section [2].

^{14}C was produced via the $^{11}\text{B}(^4\text{He}, \text{p})$ reaction by irradiating boron powder with a 3 MeV ^4He beam from the 1 MV Tandetron accelerator. The boron powder was packed into a cathode holder with a diameter of 1.5 mm, which is used in the ion source (S5) for AMS. The irradiation time was approximately 4 h, and the beam intensity during irradiation was 4–12 nA. Taking into account that the cathode holder diameter was smaller than the beam diameter, the number of produced ^{14}C atoms was estimated to be 2.1×10^7 from the integrated charge of the ^4He beam and nuclear data [3]. The cathode holder containing the irradiated boron powder was then loaded into the S5 of the 6 MV Pelletron tandem accelerator. After optimizing the beamline under the same beam conditions as those used for ^{14}C -AMS measurements, ^{14}C was detected using the ionization chamber (IC) at the end of the L4 beamline.

Figure 1 shows the time dependence of the ^{14}C counts measured by the IC. The count rate reached its maximum at approximately 9 min and decreased to half of the maximum value at approximately 17 min after the start of sputtering. Compared with the case of a ^{14}C standard sample [4], the count rate decreased more rapidly. Based on the range of the ^4He beam, this behavior is attributed to the fact that ^{14}C was produced only within a region extending to a depth of 13.0 μm from the sample surface, and that this region was removed by sputtering within a short time. A total of ~ 3000 ^{14}C atoms were detected by the IC over 40 min, and the extraction efficiency at the end of the beamline was found to be approximately 0.01%. In future work, we aim to improve the ^{14}C count rate by increasing the energy and intensity of the ^4He beam.

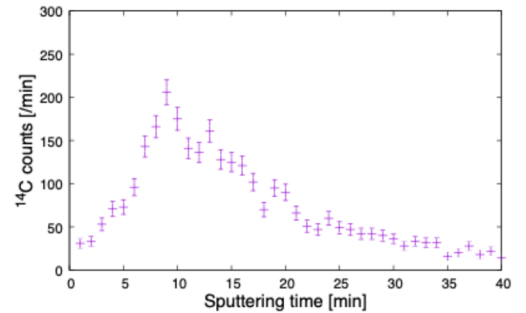


Fig. 1. Count rate of ^{14}C , produced via the $^{11}\text{B}(^4\text{He}, \text{p})$ reaction, measured by the IC as a function of sputtering time.

References

- [1] M. Mikawa et al., UTTAC Annual Report 2024, UTTAC-94 (2025) 3.
- [2] Y. Jiang et al., *Astrophys. J.* 989 (2025) 231.
- [3] R. A. Dayras et al., *Nucl. Phys. A* 261 (1976) 365.
- [4] M. Mikawa et al., this Annual Report 2.4.

2.4 Time dependence of count rates for ^{10}Be and ^{14}C beams

M. Mikawa, T. Moriguchi, A. Ozawa, K. Sasa, T. Takahashi, M. Matsumura, T. Yoshida, S. Ishii, K. Ishimoto, T. Manako, A. Yano, M. Mitsui, H. Kobayashi, R. Iwamoto, Y. Saito

Accelerator Mass Spectrometry (AMS) is typically used to measure the isotopic ratios of long-lived nuclides (^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{129}I , etc.). In FY2024, we started to develop radioisotope (RI) beams of ^{10}Be and ^{14}C , which were extracted from the AMS system for nuclear physics experiments [1]. Using the AMS system, RI beams can be extracted directly from the ion source. Therefore, an advantage of the present method is that the energy distribution is narrower than that of RI beams produced via nuclear reactions. In FY2025, the time dependence of the count rates was investigated for ^{10}Be and ^{14}C beams.

The AMS standard samples (isotope ratios of $^{10}\text{Be}/\text{Be} \sim 10^{-11}$ and $^{14}\text{C}/\text{C} \sim 10^{-12}$) were loaded into sample holders (cathodes) with a diameter of 1 mm. Sputtering was performed sequentially on four ^{10}Be samples and three ^{14}C samples, and the total counts were measured in successive 3-minute intervals by an ionization chamber located at the end of the L4 course. Figure 1 shows the time dependence of the count rates. Solid lines and their bands indicate the averages and standard deviations for ^{10}Be and ^{14}C beams.

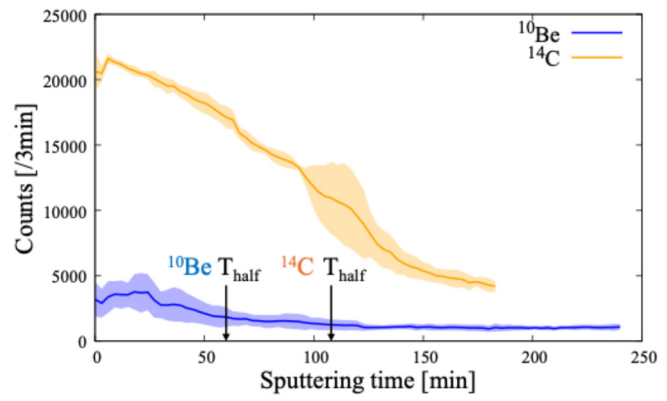


Fig. 1. The time dependence of the count rates is shown in blue for ^{10}Be and in orange for ^{14}C .

The elapsed time from the start of sputtering until the counts decreased to half of the maximum (T_{half}) was approximately 58 minutes for ^{10}Be and 111 minutes for ^{14}C . Assuming that 39 standard samples of ^{10}Be or ^{14}C are fully loaded into the cathode holder of the S5 ion source [2], and that each sample is used for a duration of T_{half} , the continuous supply time is estimated to be 39 hours (8×10^6 counts) for the ^{10}Be beam and 72 hours (2×10^7 counts) for the ^{14}C beam without replacing the cathode holder. Further improvement of the count rates is planned.

References

- [1] M. Mikawa et al., UTTAC Annual Report 2024, UTTAC-94 (2025) 8-9.
- [2] K. Sasa et al., Nucl. Instr. Meth. Phys. Res. B 361, (2015) 124-128.

2.5 Development of a radioactive ^7Be beam

Y. Saito, T. Moriguchi, A. Ozawa, K. Sasa, T. Takahashi, M. Matsumura, T. Yoshida, S. Ishii, K. Ishimoto, T. Manako, A. Yano, M. Mikawa, M. Mitsui, H. Kobayashi, R. Iwamoto

The abundance of ^7Li predicted by big-bang nucleosynthesis (BBN) is about three times larger than the observed value [1]. Since ^7Li is mainly produced by the electron-capture decay of ^7Be after BBN, reaction mechanisms that reduce the amount of ^7Be are important for the cosmological lithium problem. The purpose of this study is to develop a low-energy radioactive ^7Be beam at UTTAC for studies of nuclear reactions related to the BBN Gamow window of 0.03–1 MeV.

To produce ^7Be via the $^7\text{Li}(p, n)$ reaction, whose threshold energy is 1.88 MeV, Li_2O powder mixed with Ag powder, as a target sample, was irradiated with a 2.0 MeV proton beam from the 1MV Tandetron accelerator. The irradiation time was approximately 4 h with a typical beam current of 100 nA. The sample was then installed in the MC-SNICS ion source, S5, of the 6MV tandem accelerator. A $^7\text{Be}^{4+}$ beam was accelerated to 25.8 MeV and transported to an ionization chamber (IC) at the end of the L4 beamline. The IC signals were calibrated using ^7Li and ^{10}Be beams at 19.8 MeV and 20.3 MeV, respectively. Fig. 1 shows the particle identification using two energy-loss signals ($\Delta E1$ and $\Delta E2$) from the IC. As shown in Fig. 1, we successfully identified ^7Be particles. The count rate of ^7Be reached a maximum of 42 cpm and decreased to half of its maximum value 7.8 min after the start of sputtering. The beam transport of ^7Be could not be optimized due to the rapid decrease in the count rate. A total of $\sim 4 \times 10^2$ ^7Be particles ($N_{\text{det.}}$) were counted in 30.5 min by the IC.

Gamma-ray spectra of the Li_2O sample were measured with a Ge semiconductor detector before and after the extraction of a ^7Be beam, as shown in Fig. 2. The 477.6 keV peak corresponding to the decay of ^7Be was clearly observed before extraction, whereas it almost disappeared after extraction. From the difference between the red and blue peaks in Fig. 2, the number of ^7Be particles extracted from the sample (ΔN) was estimated to be $\sim 1.5 \times 10^{10}$. The extraction efficiency ($N_{\text{det.}}/\Delta N$) was determined to be $\sim 3 \times 10^{-8}$. These results demonstrate the feasibility of radioactive ^7Be beam production at UTTAC, although further improvement of the beam intensity is required for future reaction studies.

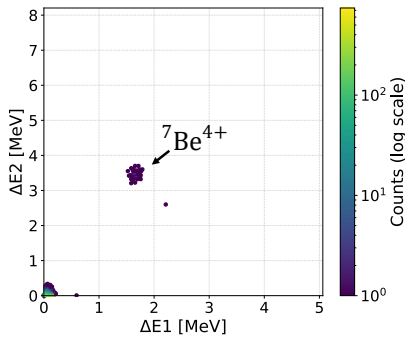


Fig. 1. Particle identification of a ^7Be beam with the IC.

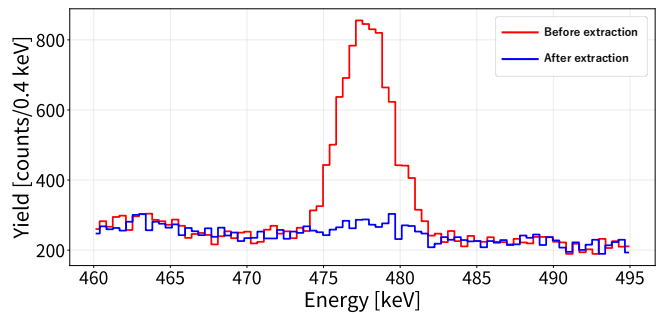


Fig. 2. Gamma-ray spectra of the Li_2O sample before (red) and after (blue) the extraction of a ^7Be beam.

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2.6 Investigation of the relationship between emission wavelength in inorganic scintillators and incident particle type and energy

T. Iida, M. Yoshino¹, T. Moriguchi, A. Ozawa, M. Mikawa, M. Mitsui, H. Kobayashi, R. Iwamoto, Y. Saito

Scintillators are materials that convert radiation into visible light. Recent studies by our research group have revealed that some scintillator materials exhibit changes in scintillation emission wavelength depending on the type of incident particle. Conventional particle identification methods using scintillators have mainly relied on scintillation decay time information; however, the additional use of emission wavelength information may enable new approaches to particle discrimination and contribute to the development of next-generation particle detectors. The purpose of this study is to investigate how incident particle species and their energies affect the luminescence properties of various scintillator materials by irradiating them with multiple charged particles of different atomic numbers Z and energies using the 6MV tandem accelerator at UTTAC. One example is the Eu-doped CaF_2 scintillator. When a small amount of Eu is doped into CaF_2 , Eu^{2+} ions substitute for Ca^{2+} sites, producing blue emission originating from the Eu^{2+} 5d-4f transition. On the other hand, increasing the Eu concentration allows Eu^{3+} ions to occupy part of the sites in the CaF_2 crystal lattice, resulting in additional red emission originating from Eu^{3+} 4f-4f transitions simultaneously with the Eu^{2+} emission [1]. In our previous study, we found that the relative intensity ratio of Eu^{2+} and Eu^{3+} emission changes depending on the type of incident radiation [2].

To investigate this radiation-type dependence of Eu: CaF_2 luminescence in detail, we initiated tandem accelerator experiments in FY2025. In the experiment, scintillator crystals were placed inside a vacuum chamber on the A7 beamline and irradiated with various ion beams. The scintillation light was transmitted outside the chamber through an optical fiber installed inside the vacuum chamber, and the luminescence spectra were measured using an optical spectrometer. Figure 1 shows the spectra obtained from a Eu 3%: CaF_2 crystal irradiated with five different radiation types. The horizontal axis represents wavelength (nm), while the vertical axis shows relative intensity normalized such that the Eu^{2+} peak at 420 nm is approximately 100. It can be seen that the Eu^{3+} -related emission on the longer-wavelength side increases for heavier ions with larger dE/dx .

In the future, we will investigate the dependence of the emission wavelength on the incident particle energy, in addition to the particle species.

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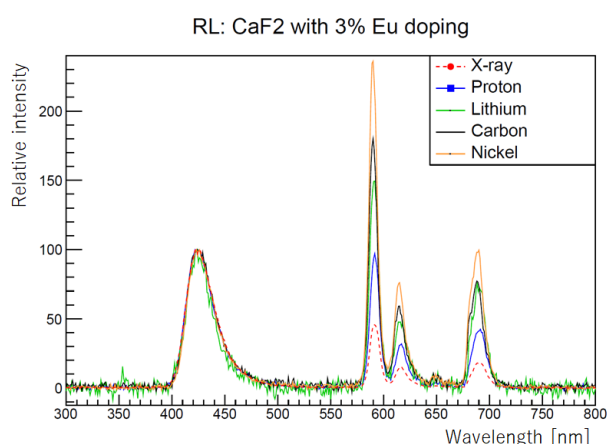


Fig. 1. Luminescence spectra of CaF_2 crystals doped with 3% Eu when irradiated with five different types of radiation particles.

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2.7 Investigation of ions ejected from an arc plasma ion source

A. Ozawa, H. Kobayashi, M. Hayashi, T. Moriguchi, A. Moriyama, Y. Seki, Y. Yano

The Rare RI Ring (R3), located in the RI Beam Factory at RIKEN, is a storage ring for mass measurements of rare RIs based on isochronous mass spectrometry [1]. To tune the isochronous magnetic field in R3, beams from an accelerator are usually necessary. If we can use a beam from a standalone ion source located in R3, we can tune the isochronous magnetic field offline. This gives benefits to R3, i.e., a reduction of the tuning time in mass measurements and the production of a more precise isochronous field. Thus, we began to search for an ion source that can be used in R3. In R3, the beam energy of rare RI beams is roughly 180 MeV and its rigidity is about 4.5 Tm. To obtain this high rigidity, we searched for an ion source producing atomic clusters. With $W_{n=500}$ clusters at 10 keV, an ion beam with this high rigidity can be obtained. Commercially, the following cluster ion sources are available: the evaporation ion source [2], Ar gas cluster ion source [3], and arc plasma ion source (APS) [4]. Among them, we investigated the performance of the evaporation ion source in a previous study [5]. In that study, we succeeded in producing $W_{n=100}$ cluster ions at 1 keV. However, the intensity of the cluster ions was quite low (~ 0.2 cps) [5]. It is difficult to apply this ion source to R3. According to Ref. [3], the intensity of $Ar_{n=3000}$ gas clusters seems to be sufficiently high (more than 500 nA). $Ar_{n=3000}$ gas cluster ions at 10 kV are available, which corresponds to a rigidity of 5.0 Tm. However, Ar gas clusters may evaporate immediately at room temperature ($T \sim 300$ K). In R3, clusters should survive for at least one turn to tune the isochronous field, which corresponds to 15 ms. Thus, from the viewpoint of lifetime, the Ar gas cluster ion source cannot be used in R3, and metallic clusters (W, Cu, Al) are indispensable. The APS can produce metallic clusters. Since the APS has been used for film formation, it seems to achieve sufficient intensity for clusters. The APS has another merit: since only an arc plasma is needed, the APS can be installed even in high vacuum. Thus, we can install the APS in R3 without a differential pumping system. However, cluster ions from the APS have not been directly observed so far. Thus, in the present study, we tried to observe cluster ions from the APS directly.

The experiment was performed at the A7 course of UTTAC. We located the APS at the end of the straight line of A7 and installed Cu material at the head of the APS. To detect ions from the APS, we located a microchannel plate (MCP) inside the entrance chamber of A7. The distance from the APS to the MCP was roughly 2.4 m. In the APS, a trigger signal for the arc is available. This signal can be used as a start signal for the time of flight (ToF). Thus, we can measure the ToF between the APS and the MCP. We also installed a mesh biased up to ± 5 kV and a shutter. Downstream of the entrance chamber of A7, there is a bending magnet where a few hundred G can be applied.

To obtain signals of positive ions from the APS, we applied -2.4 kV to the MCP. The MCP signal was shaped by a discriminator, and the ToF between the APS and the MCP was measured by a long-range TDC (P7887, FAST Comtec GmbH). Before the measurements using the APS, we observed signals of alpha particles from ^{241}Am with the MCP. Since an arc plasma is generated in the APS, blue plasma light can be observed. Since the MCP is sensitive to this light, it gives a time reference for the ToF. According to Ref. [4], at the head of the APS, an arc voltage of ~ 10 kV is applied. If $Cu_{n=1000}$ cluster ions with 10 keV are

produced, a ToF signal is expected at ~ 0.5 ms.

A typical observed ToF spectrum is shown in Fig. 1. The sharp peak located near $7\ \mu\text{s}$ is the signal from the plasma light in the APS. The sharp peak located near $100\ \mu\text{s}$ is related to the trigger signal. We could not observe any sharp peak near 0.5 ms, although a broad peak was observed in the range of 1 – 10 ms. All peaks disappeared when we put the shutter between the APS and the MCP. If ion signals were included in this broad peak, they would be affected by the mesh voltage and the magnetic field of the bending magnet. However, the broad peak was not affected by changing the mesh voltage or the magnetic field. Thus, we conclude that the observed broad peak does not originate from ions coming directly from the APS. After the experiments, a Cu film was created on the inner surface of the exit duct, and the Cu material was chipped. Thus, some material was actually ejected from the APS. However, we did not observe with the MCP any ion-like signal coming directly from the APS. Thus, we conclude that no energetic ions are ejected from the APS. In the APS, an arc plasma is created, and the temperature must be locally high enough to evaporate the material. Thus, evaporation must occur, and large amounts of neutral evaporation residues are ejected from the APS. The broad peak in Fig. 1 may be related to these neutral evaporation residues. When these neutral residues collide with the vacuum duct, secondary electrons may be created. These secondary electrons may be the source of the observed broad peak. These secondary electrons are not affected by the mesh voltage or the magnetic field. There is a possibility that low-energy ions, which cannot be measured with our MCP, are produced. Since the MCP is a standard ion detector, ions that cannot be detected with the MCP are not applicable in R3. In conclusion, the APS cannot be used to tune the isochronous magnetic field in R3.

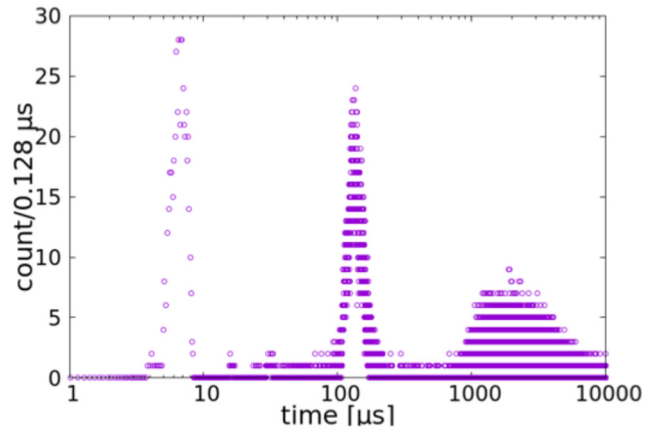


Fig. 1. Typical ToF spectrum observed using Cu material in the APS.

The authors thank ADVANCE RIKO, Inc., which lent us the APS ion source and gave us technical suggestions. The authors also thank Prof. S. Tomita, who lent us the long-range TDC.

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3.

ACCELERATOR MASS SPECTROMETRY

3.1 Operational status of the Tsukuba 6 MV multi-nuclide AMS system in FY2025

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The Tsukuba 6 MV multi-nuclide AMS system was operated for a total of 51 days (613.3 hours) in FY2025. We conducted 9 research projects related to AMS. Figure 1 shows the monthly numbers of measured radionuclide samples from April 2025 to March 2026. During this period, 679 samples were measured, mainly for ³⁶Cl and ¹²⁹I. Beam production tests of ¹⁰Be and ¹⁴C radioactive ion beams were carried out [1]. In addition, ²¹⁰Pb-AMS was under development in FY2025 [2]. Table 1 shows the breakdown of the measured samples. Table 2 summarizes the AMS measurement conditions for each radionuclide. Background (BKG) isotope ratios ranged from 10⁻¹⁵ to 10⁻¹⁴ except for ²¹⁰Pb. The problem with the sample rotation mechanism of the MC-SNICS ion source, which arose in FY2024, has not been resolved, and the sample wheel can still rotate only in one direction.

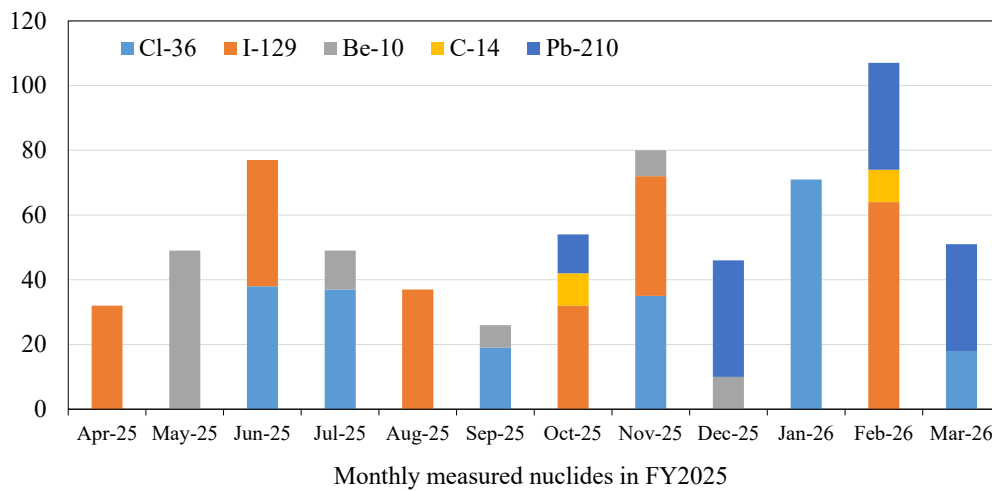


Fig. 1. Monthly numbers of measured radionuclide samples from April 2025 to March 2026.

Table 1. Number of measured samples in FY2025.

Nuclides	Reagents and blanks	Standard samples	Measured samples	Total
Be-10	24	39	23	86
C-14	0	16	4	20
Cl-36	50	55	113	218
I-129	47	21	173	241
Pb-210	54	60	0	114
Subtotal	175	191	313	679

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Table 2. AMS measurement conditions for radionuclides in FY2025.

Nuclides	Terminal voltage [MV]	Accelerated ion	Total energy [MeV]	Beam current (Low Energy side F.C.) [μ A]	Transmission [%]	Measured/ Known [%]	BKG [atomic ratio]
Be-10	6.0	$^{10}\text{Be}^{3+}$	20.3	1.5–3	15–20	75	10^{-15}
C-14	5.0	$^{14}\text{C}^{4+}$	25.0	0.5–50	26–40	–	–
Cl-36	6.0	$^{36}\text{Cl}^{7+}$	48.0	10–15	8–15	70	10^{-15}
I-129	5.0	$^{129}\text{I}^{5+}$	30.0	0.5–4	5–10	90–98	10^{-14}
Pb-210	3.0	$^{210}\text{Pb}^{5+}$	17.6	~ 0.1	–	–	–

^{36}Cl -AMS

^{36}Cl -AMS has been performed under the beam condition of $^{36}\text{Cl}^{7+}$ with 48.0 MeV at the terminal voltage of 6 MV (Table 2). In FY2025, we mainly conducted ^{36}Cl -AMS to investigate fluctuations in cosmic ray intensity in Antarctic NDF ice cores in collaboration with Nagoya University and Hirosaki University [3]. We also conducted an experiment to estimate the accumulated thermal neutron dose using ^{36}Cl at the CERN/CHARM experimental facility in collaboration with KEK [4].

^{129}I -AMS

^{129}I -AMS has been performed under the beam condition of $^{129}\text{I}^{5+}$ with 30.0 MeV at the terminal voltage of 5 MV (Table 2). In FY2025, ^{129}I -AMS was applied to tracer studies investigating the environmental dynamics of ^{129}I in river and sea waters in collaboration with Kanazawa University and Fukushima University. Detailed ^{129}I measurement results are summarized in Ref. [5].

Measurement history

Figure 2 shows the annual numbers of measured samples since the Tsukuba 6 MV multi-nuclide AMS system started operation. A total of 6,589 samples were measured from FY2016 to FY2025.

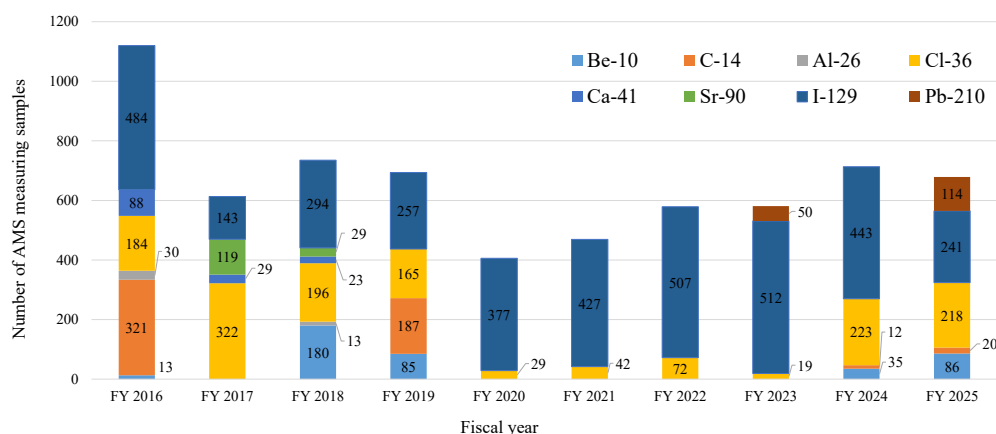


Fig. 2. Annual numbers of AMS measured samples from FY2016 to FY2025.

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3.2 The Performance of iodine-129 AMS measurements at the University of Tsukuba (FY2025)

M. Matsumura, T. Yoshida, K. Sasa, T. Takahashi, T. Matsunaka¹

The performance of ^{129}I measurements by Accelerator Mass Spectrometry (^{129}I -AMS) has been reported since 2017 [1]. In FY2025, we measured 241 samples prepared as AgI targets, which is 35.5% of all the measured AMS samples. The details are shown in Fig. 1 and described also in this annual report by K. Sasa et al. [2]. Figure 2 shows the measured blank-values of AgI precipitated from carrier-reagent, called old iodine, and the ^{127}I beam current measured by the MFC02-2 Faraday cup before injection to the accelerator, average for each fiscal year. The average of the $^{129}\text{I}/^{127}\text{I}$ ratios was $(2.7 \pm 2.2) \times 10^{-14}$ in FY2025. The average of ^{127}I beam current at the MFC02-2 cup was 3.2 μA in FY2016–2017 and decreased year by year. However, contrary to such a trend, it seems to have increased slightly to 1.1 μA in FY2025.

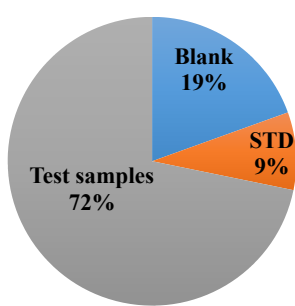


Fig. 1. Measured samples by ^{129}I AMS in FY2025.

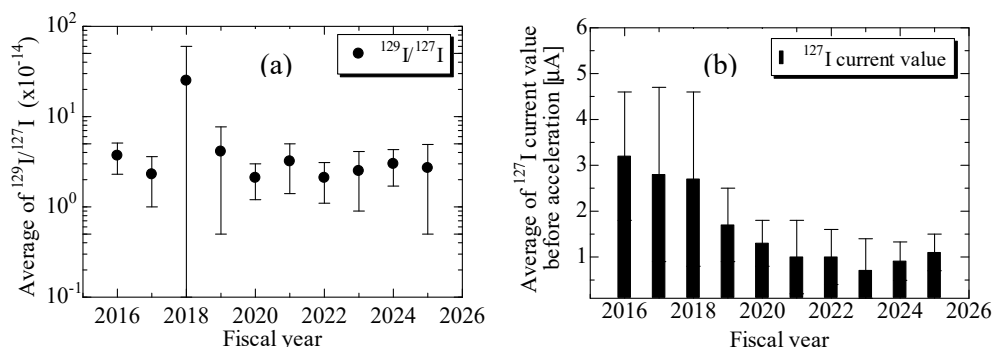


Fig. 2. The ratio of $^{129}\text{I}/^{127}\text{I}$ of old iodine blank values (a) and ^{127}I beam current (b), measured since FY2016.

The measured $^{129}\text{I}/^{127}\text{I}$ values were normalized by using Standard Reference Material (SRM), Purdue-1 STD (Z94-0597) and Purdue-2 STD (Z94-0596) with a $^{129}\text{I}/^{127}\text{I}$ ratio of 8.378×10^{-12} and 6.540×10^{-11} [3] (the value was revised in 2014 [4]), which was provided by the Purdue Rare Isotope Measurement Laboratory (PRIME Lab) at Purdue University, USA. The averaged $^{129}\text{I}/^{127}\text{I}$ ratios with their standard deviations (1σ) are $(7.227 \pm 0.270) \times 10^{-12}$ (3.7%) and $(5.785 \pm 0.369) \times 10^{-11}$ (6.4%) for Purdue-1 ($n = 165$) and Purdue-2 ($n = 138$), respectively, in FY2025. The ratios of the measured average $^{129}\text{I}/^{127}\text{I}$ ratio to the nominal ratio are 0.8626 ± 0.0322 and 0.8846 ± 0.0564 for Purdue-1 and Purdue-2, respectively, which are consistent within the margin of error. The ^{127}I beam currents for Purdue-1 and Purdue-2 in FY2025 were 1.6 μA and 1.8 μA , with transmission rates before and after acceleration of 10% and 9%, respectively.

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3.3 Survey of iodine-129 isotope ratios in groundwater in the Hokuriku Region

M. Matsumura, T. Matsunaka¹, T. Yoshida, T. Takahashi, K. Sasa

In this study, we collected hot spring water from the Wakura region and natural groundwater from spring sites at several locations on both the mountain and sea sides centered on Kanazawa City, Hakui City, Kahoku District, and Hakusan City along the Ouchigata Fault Zone and the Morimoto-Togashi Fault Zone, as well as upstream river water from the Asano River, and measured the iodine isotope ratio ($^{129}\text{I}/^{127}\text{I}$), where ^{129}I is present at trace levels. This report presents the measurement and analysis results for spring water samples collected in FY2024 and analyzed in FY2025.

Analyses were conducted on groundwater collected from 13 locations in March, May, June, and November 2024. The ^{129}I concentrations in the groundwater ranged widely from $(0.023\text{--}22) \times 10^{-6} \text{ mBq L}^{-1}$ with a mean value of $2.9 \times 10^{-6} \text{ mBq L}^{-1}$. Relatively low values were observed at "Mitarashi-ike Pond," which is at the northern end of the sampling area, at $(0.14\text{--}1.2) \times 10^{-6} \text{ mBq L}^{-1}$, and at the nearby "Kobo-no-reisen Spring," at $(0.023\text{--}2.5) \times 10^{-6} \text{ mBq L}^{-1}$; even at the same site, concentrations varied depending on the sampling period. The highest value was found at "Sakurashozu" near the southern end of the sampling area, with a concentration of $2.2 \times 10^{-5} \text{ mBq L}^{-1}$ in the sample collected in June 2024. The stable iodine concentration ranged from $(0.43\text{--}18) \mu\text{g L}^{-1}$ across all groundwater samples. The minimum value was at "Yuwaku-no-meisui Spring," located on the mountain side of Kanazawa City in the sample collected in November 2024, and the highest value was at "Sakurashozu," collected in June 2024. The mean value was $3.1 \mu\text{g L}^{-1}$. The $^{129}\text{I}/^{127}\text{I}$ ratio also ranged widely from $(0.017\text{--}7.8) \times 10^{-10}$ in all groundwater samples. Relatively low values were observed at "Mitarashi-ike Pond" at the northern end, at $(1.2\text{--}8.5) \times 10^{-11}$ and "Kobo-no-reisen Spring" at $(0.17\text{--}18) \times 10^{-11}$, varying by sampling period, similar to the ^{129}I concentrations. The highest value, 7.8×10^{-10} , was found at the "Ohama-no-mizu Spring" of the Hakusan-Mikawa Subsurface Water Group, sampled in June 2024, at a site near its southern end. Overall, the mean $^{129}\text{I}/^{127}\text{I}$ ratio was 1.6×10^{-10} , and the ratios were higher in groundwater in the southern part of Ishikawa Prefecture than in the northern part. Going forward, to examine whether the "Hakusan-Mikawa Subsurface Water Group" shows characteristically high $^{129}\text{I}/^{127}\text{I}$ values, it will be necessary to conduct a continuous and comprehensive survey by expanding the sampling sites along faults in Ishikawa Prefecture to both the sea and mountain sides, as well as further north and south. In addition, we plan to compare these results with those for hot spring water and river water.

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3.4 Test measurements of ^{10}Be in Antarctic ice core samples

H. Takami, K. Sasa, M. Matsumura, T. Yoshida, T. Takahashi, F. Miyake¹, K. Horiuchi²

When cosmic rays enter the Earth's atmosphere, they induce a chain of nuclear reactions that produce radionuclides known as cosmogenic nuclides, such as ^{10}Be , ^{14}C and ^{36}Cl . Large cosmic-ray events can produce peaks in the concentrations of cosmogenic nuclides. To investigate the scale of cosmic ray events and the origin of the rays, it is necessary to measure various nuclides. This is because the production rate of cosmogenic nuclides depends on the type, energy, and intensity of the incoming cosmic rays. Until now, we had only measured ^{36}Cl in ice cores to investigate cosmic ray events; however, recognizing the need for further analysis, we conducted a pilot measurement of ^{10}Be in the NDF2018 ice core in this study, at the University of Tsukuba Tandem Accelerator Complex (UTTAC).

In this study, we measured $^{10}\text{Be}/^9\text{Be}$ ratios in 14 sections of NDF2018 Antarctic ice core samples (depth: 49.245–49.745 m) and some standards, using the AMS instrument at UTTAC. The measurement sequence took approximately 42 hours to complete.

The maximum extracted $^9\text{BeO}^-$ beam current was approximately 2.5 μA . This value is significantly lower than that reported in a previous study conducted at UTTAC in 2019 [1]. Figure 1 shows measured $^{10}\text{Be}/^9\text{Be}$ ratios of the standard samples. The variation throughout the entire measurement sequence was 3.8%. Instability in the standard sample measurements was observed during the initial stage of the measurement sequence.

Based on these results, we plan to address the following challenges moving forward: increasing the BeO^- beam current and improving the reproducibility of standard sample measurements.

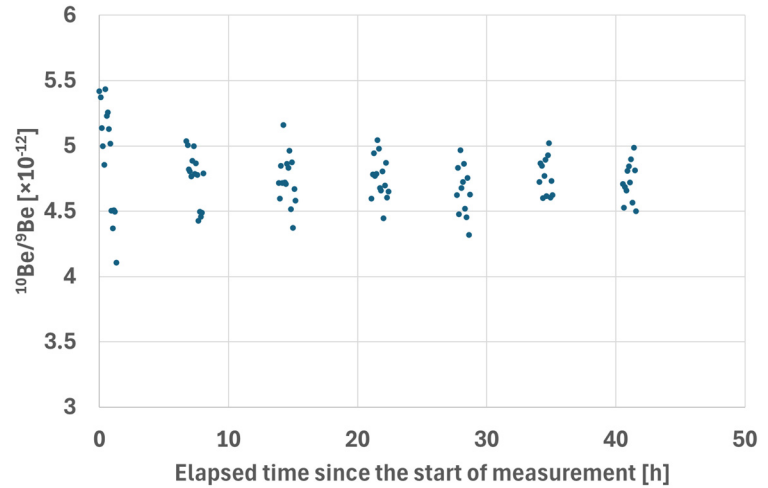


Fig. 1. Measured $^{10}\text{Be}/^9\text{Be}$ ratios of the standard samples. The ratios were unstable during the initial stage of the measurement sequence.

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3.5 Study of ^{210}Pb measurement using Accelerator Mass Spectrometry

M. Sakuma, A. Sakaguchi, M. Matsumura, T. Takahashi, T. Yoshida, K. Sasa

The radionuclide ^{210}Pb , with a half-life of 22.2 years, belongs to the ^{238}U decay series and is deposited in soil and sediment. Due to its relatively short half-life, it is suitable for dating samples from the past ~100 years and has attracted attention as an indicator radionuclide for environmental samples from the Anthropocene [1]. Conventional ^{210}Pb measurements have relied on radiation counting and ICP-MS techniques, but these approaches face challenges such as sample volume, measurement time, sensitivity, and isobaric interference [2]. Therefore, in this study, we investigated the measurement conditions to develop a highly sensitive and accurate ^{210}Pb measurement method using the 6 MV tandem accelerator AMS system at UTTAC.

PbO_2^- ions were extracted from Pb_3O_4 targets mixed with metal powder. When Ag and Au were compared as mixing metals, high ion currents were obtained under the conditions of Pb sample: Ag = 1:1 and Pb sample: Au = 2:1 (mass ratio), and for Ag mixing, a stable beam current was maintained. Figure 1 shows the transmission as a function of acceleration voltages and Pb ion charge states. High transmission was obtained at an acceleration voltage of 2 MV with a charge state of 4+ (9.7 MeV), and at 3 MV with a charge state of 5+ (17.6 MeV), which were generally consistent with previous studies [3]. A gas ionization chamber was used as the detector, and based on PHITS simulations [4], a 75 nm Si_3N_4 film was used as the detector window, and 10 Torr of isobutane was used as the detector gas.

Currently, we are continuing test measurements using blank and standard samples with $^{210}\text{Pb}/^{208}\text{Pb}$ ratios ranging from 10^{-9} to 10^{-11} . Although the measured spectra showed variations corresponding to the isotopic ratios, non-negligible counts were also observed in the blank measurements, suggesting interference from the stable isotope ^{208}Pb . Furthermore, a method for measuring the ^{208}Pb beam current using the high energy offset Faraday cup has not yet been established and remains a challenge for quantitative evaluation of isotopic ratios. We will continue to re-examine the detection conditions and conduct test measurements with the aim of applying this method to actual samples.

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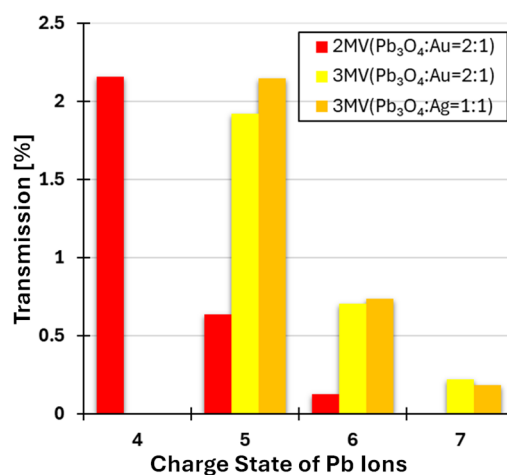


Fig. 1. Dependence of transmission on acceleration voltages and Pb ion charge states

3.6 AMS Measurements of $^{36}\text{Cl}/^{35}\text{Cl}$ Ratio in Cable Sheaths Irradiated at the CERN CHARM Facility

T. Kimura, G. Yoshida¹, H. Matsumura¹, M. Matsumura, T. Sanami¹, M. Ishida¹, R. Shiobara², M. Mitsuhashi², H. Yashima³, T. Yoshida, T. Takahashi, K. Sasa

At high-energy accelerator facilities, various components such as concrete, electromagnets, and cables are activated by secondary particles generated by beam loss. These components then become radioactive waste when the facility is decommissioned, resulting in a significant waste-management problem. In the main ring tunnel of the 12 GeV Large Proton Synchrotron (12GeV-PS) at the High Energy Accelerator Research Organization (KEK), which operated from 1976 to 2005, widespread activation of concrete and electromagnets has been observed. The effect is especially pronounced at EP1 and EP2, which correspond to branching points of the beamline [1]. Cables, which are one type of surrounding components, are found throughout accelerator facilities, but their activation has not been a focus of attention. For example, the cables around EP1 consist of twisted thin copper wires (conductors) covered with a sheath composed mainly of chlorinated polyethylene; various nuclides are generated in them by secondary particles caused by beam loss. In particular, the long-lived nuclide ^{36}Cl (half-life: 301,000 years), produced by the neutron capture reaction of the stable isotope ^{35}Cl contained in the sheath, almost never decays and can be a source of residual radioactivity over the long term. A previous study has quantified ^{36}Cl in the EP1 sheath [2]. In the future, it is necessary to conduct a detailed investigation of ^{36}Cl in sheaths. In this study, irradiation experiments were conducted at the CERN High-energy Accelerator Mixed field facility (CHARM) using secondary neutrons generated by beam loss to acquire basic data on the production of ^{36}Cl in sheaths. ^{36}Cl was quantified by Accelerator Mass Spectrometry (AMS) using the 6 MV tandem accelerator at the University of Tsukuba, which has a wealth of experience in measuring this isotope.

The following is an overview of the experiment. A new cable, similar to those used in the 12GeV-PS, was purchased and used as the sample. Prior neutron activation analysis using Kyoto University Reactor (KUR) estimated the chlorine concentration in this sheath to be approximately 10 wt%. The floor plan of the CHARM facility is shown in Figure 1. This facility uses a 24 GeV/c proton beam to impinge on a copper target, providing a neutron field covering wide energy ranges. Samples cut to approximately 5 cm in length were placed at five different positions from A to E in the CHARM facility and irradiated for about 60 hours. In this experiment, the average beam intensity was $(4.40 \pm 0.82) \times 10^{10}$ proton s^{-1} . After irradiation, a portion of the sheath was collected from each sample. Approximately 0.3 g of sheath was combusted, and the generated gas was collected using two stages of trapping water to extract chlorine as chloride ions. Ion chromatography was then performed on the water from the first trap to measure Cl^- and SO_4^{2-} (an interfering factor for AMS), and the water was diluted to an isotope ratio suitable for AMS measurement ($^{36}\text{Cl}/^{35}\text{Cl} \sim 5 \times 10^{-12}$) by adding a chlorine carrier. Chemical operations, such as the addition of AgNO_3 , were then

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performed to purify the AgCl precipitate as samples for AMS. Chlorine ions (Cl^-) were extracted from AgCl set in a Cs sputter negative ion source and injected into the 6 MV tandem accelerator, where the acceleration voltage was raised to 6 MV. They were converted to positive ions by a carbon stripper foil ($4.8 \mu\text{g cm}^{-2}$) at the terminal and accelerated again. After the target ions ($^{36}\text{Cl}^{7+}$, 48 MeV) were identified through analysis using magnetic and electrostatic fields, they were detected by a gas ionization detector [3] filled with isobutane gas at 27 Torr. Measurements were taken for 180 to 240 seconds per sample.

The results of the $^{36}\text{Cl}/^{35}\text{Cl}$ ratio are shown in Figure 2. The $^{36}\text{Cl}/^{35}\text{Cl}$ ratios in the irradiated sheaths at CHARM ranged from approximately 10^{-12} to 10^{-10} , achieving sufficient production of ^{36}Cl for AMS measurements. This opens the door to various experimental configurations, making it possible to conduct detailed investigations into ^{36}Cl production in sheaths and the activation of cables.

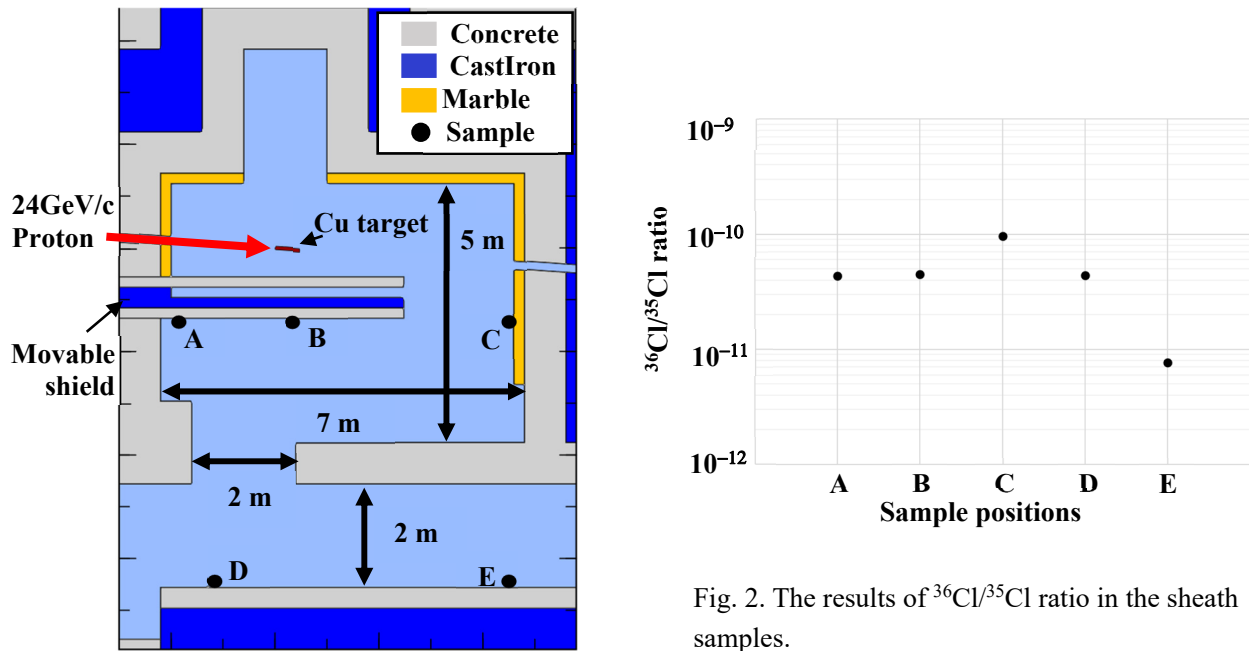


Fig. 1. The floor plan of the CERN CHARM facility. The black dots indicate the positions of the samples.

Fig. 2. The results of $^{36}\text{Cl}/^{35}\text{Cl}$ ratio in the sheath samples.

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3.7 Riverine ^{129}I dynamics in the Hamadori rivers

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The long-lived ^{129}I is one of radionuclides released by the Fukushima Daiichi Nuclear Power Plant accident. To evaluate redistribution in the terrestrial environment and influence on the marine environment of ^{129}I , we continued analyses of ^{129}I in river waters collected from the Abukuma and Niida rivers in a framework of the Environmental Radioactivity Research Network Center (ERAN) program^[1]. In fiscal year 2025, we analyzed ^{129}I concentrations in river water samples collected during high-flow events at three rivers flowing through the Hamadori area which is assumed to be exposed to substantial ^{129}I deposition.

Water samples of 20 L were collected at downstream points on the Niida, Ukedo and Takase rivers on 4, 6 and 9 September 2023. Respective catchment areas at the sampling points were 206, 143 and 259 km². Catchment mean rainfall on the days ranged from 42 to 109 mm. All river water samples were filtered, and the obtained samples of suspended solids (SS) and filtrates were measured for ^{137}Cs concentrations. A portion of samples of SS and filtered water was measured for $^{129}\text{I}/^{127}\text{I}$ ratio with accelerator mass spectrometry, and measured for ^{127}I concentration with ICP-QQQ-MS after pretreatment of pyrohydrolysis procedure^[1].

Table 1 shows results of the analyses. Mean concentrations of dissolved ^{129}I in the Niida, Ukedo, and Takase rivers were 0.20, 1.2, and 0.78 $\mu\text{Bq/L}$, respectively. Respective mean ^{129}I concentrations in SS were 2.9, 15, and 6.3 mBq/kg. These magnitude relations presumably reflected the magnitude of ^{129}I deposition in the catchments. Respective mean apparent K_d in the Niida, Ukedo, and Takase rivers were 14000, 13000, and 9300 L/kg and no significant difference was found among the rivers. Although the previous study suggested dependence of apparent K_d on catchment mean ^{129}I inventory^[1], it was not the case in this study. Both dissolved ^{129}I concentrations and the ^{129}I concentration in SS were positively correlated with the corresponding ^{137}Cs concentrations, suggesting similar behavior between these two radionuclides.

Table 1. Summary of results of ^{129}I analyses.

River	Event	SS conc. (mg/L)	^{129}I conc. in SS (mBq/kg)	Dissolved ^{129}I conc. ($\mu\text{Bq/L}$)	$^{129}\text{I}/^{127}\text{I}$ in SS ($\times 10^{-8}$)	$^{129}\text{I}/^{127}\text{I}$ in water ($\times 10^{-8}$)	Apparent K_d for ^{129}I (L/kg)
Niida river	2023SEP4	139	4.29	0.255	1.95	1.45	16800
	2023SEP6	1165	2.32	0.162	1.77	1.71	14300
	2023SEP8	737	2.16	0.180	2.21	2.04	12000
Ukedo river	2023SEP4	72	22.67	1.50	13.1	13.6	15100
	2023SEP6	121	13.55	0.919	9.96	9.55	14800
	2023SEP8	1479	9.32	1.20	12.6	13.5	7750
Takase river	2023SEP4	404	3.94	0.286	1.92	3.33	13800
	2023SEP6	191	9.14	1.26	11.1	12.0	7280
	2023SEP8	2663	5.68	0.794	7.87	8.35	7150

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3.8 Distribution and concentration of I-129 in seawater and rockfish off southern Fukushima

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The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident on March 11, 2011, released substantial quantities of fission products, including radioiodine and radiocesium isotopes, into the adjacent marine environments. Among these radionuclides, the long-lived isotope iodine-129 (^{129}I , half-life: 1.57×10^7 years) is of considerable environmental importance because of its long residence time, high mobility, and redox-sensitive speciation, making it a powerful tracer for anthropogenic nuclear releases. While the post-accident behavior of cesium-137 (^{137}Cs) in rockfish (*Sebastes cheni*) has been extensively monitored, showing a slow decline and elevated levels due to dietary uptake and a long biological half-life (about 190 days) [1], quantitative data on ^{129}I accumulation in marine fish off the southern Fukushima coast and its radiological implications for humans are scarce. This study aimed to clarify the precise distribution, concentration factors (CFs), and biogeochemical behavior of ^{129}I in rockfish (*Sebastes cheni*) and seawater off southern Fukushima during 2014–2015, three to four years after the FDNPP accident. Additionally, the study evaluated the $^{129}\text{I}/^{137}\text{Cs}$ and $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios to identify contamination sources and assessed the committed effective dose from ^{129}I ingestion through fish consumption.

Rockfish specimens aged 9–10 years (hatched pre-accident) were collected from three reef sites (S-1, S-2, S-3) located 5.5–6.0 km south-southeast to southeast of the FDNPP at depths of 18.4–20.0 m. These older fish were selected to capture their long-term history because they retain accident-derived radionuclides due to slow depuration. Seawater samples were collected at site S-1 in July 2014 from the near-surface (about 0 m depth), 10 m, 15 m, and 17.4 m depth layers. In situ vertical profiles of water temperature, salinity, dissolved oxygen, and turbidity were measured using a CTD profiler. Total iodine from freeze-dried rockfish tissues was extracted via a quartz-tube pyrohydrolysis-based method. The $^{129}\text{I}/^{127}\text{I}$ ratios were measured using accelerator mass spectrometry (AMS) systems at MALT (The University of Tokyo) and UTTAC (University of Tsukuba).

As illustrated by the data in Fig. 1, the coastal water column at site S-1 in July 2014 exhibited pronounced summer stratification. Water temperature decreased from 21.9 °C at the near-surface to 15.3 °C at the bottom, while salinity conversely increased from 31.8 to 33.1, developing a sharp pycnocline at approximately 15.0 m depth that restricted vertical mixing. While stable ^{127}I was nearly uniform throughout the water column with a mean of 2.74×10^{17} atoms L^{-1} (56.1 ppb), radioactive ^{129}I exhibited pronounced surface enrichment, dropping 18-fold from 158×10^7 atoms L^{-1} at the surface to about 8.7×10^7 atoms L^{-1} at 15.0–17.4 m depth. The mean ^{129}I concentration, 52×10^7 atoms L^{-1} , remained one to two orders of magnitude higher than the estimated pre-accident background level (about 1.29×10^7 atoms L^{-1} [2]).

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For rockfish, Fig. 1 displays the concentrations plotted against depth for sites S-1, S-2, and S-3. The ^{129}I concentrations in rockfish ranged from 386 to 915×10^7 atoms kg^{-1} -wet weight (5.40 – 12.8 $\mu\text{Bq kg}^{-1}$ -wet weight). The calculated ^{129}I concentration factors (CFs) were 7.4 – 18 L kg^{-1} . These CFs were approximately one order of magnitude higher than stable ^{127}I CFs (1.12 – 2.36 L kg^{-1}) but notably lower than the equilibrium CFs for stable Cs (60 – 170 L kg^{-1}). The higher CF for ^{129}I over ^{127}I is likely explained by chemical speciation, as accident-derived ^{129}I predominantly exists as iodide (I^-), which is absorbed more efficiently by fish than the iodate (IO_3^-) form dominant in stable ^{127}I . The $^{129}\text{I}/^{137}\text{Cs}$ activity ratio in the whole-body S-1 sample (2.58×10^{-7}) was in close agreement with the FDNPP reactor inventory ratio (2.95 – 3.07×10^{-7}), confirming that the radionuclides originated from the accident. Muscle samples from S-2 and S-3 exhibited lower activity ratios ($(1.21$ – $1.53) \times 10^{-7}$) due to the tissue-specific preferential accumulation of ^{137}Cs in muscle. Using the muscle tissue concentrations, the committed effective dose from ^{129}I ingestion via fish consumption was estimated to be 14 – 33 pSv year^{-1} , which is seven to eight orders of magnitude lower than the global average internal exposure from natural dietary radiation (0.29 mSv year^{-1}) [3], confirming that the radiological risk is negligible.

The integrated vertical depth profiles provided in Fig. 1 successfully verified the persistence and vertical stratification of FDNPP accident-derived ^{129}I in the coastal surface layer, which acts as a temporary reservoir slowly releasing the isotope into the North Pacific. This study provided the first field-based evidence of ^{129}I bioaccumulation in rockfish off Fukushima during the mid-term recovery phase, demonstrating that despite clear accumulation relative to stable iodine, the radiological impact on human health through fish consumption is completely negligible.

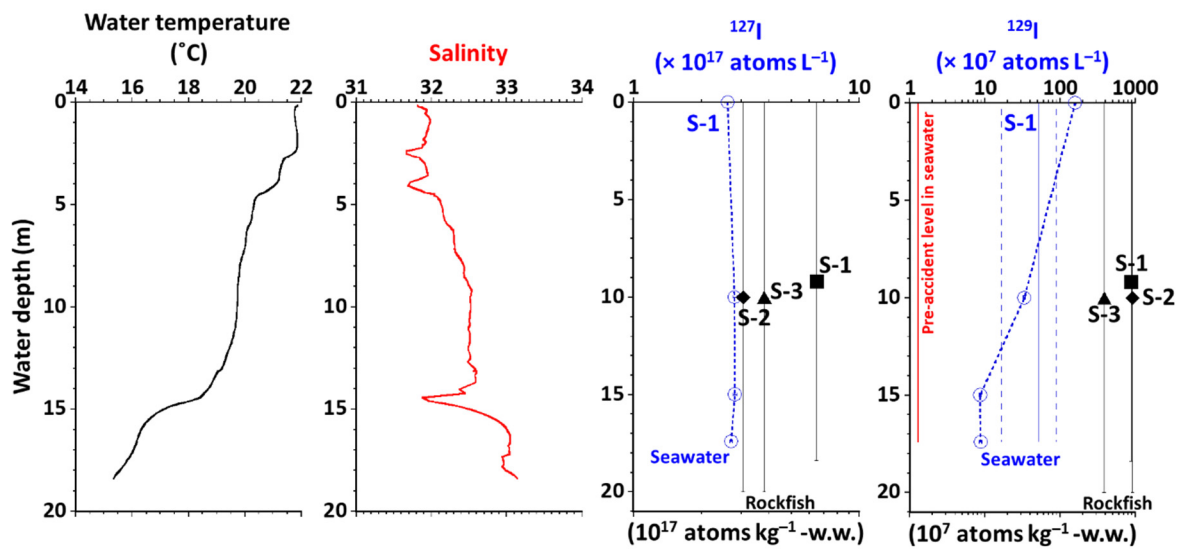


Fig. 1. Depth profiles of water temperature, salinity, and dissolved ^{127}I and ^{129}I concentrations in seawater at site S-1, along with corresponding concentrations in rockfish (*Sebastes cheni*) from sites S-1, S-2, and S-3 during 2014–2015. The red line indicates the pre-accident seawater ^{129}I background level.

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3.9 Concentrations and speciation of iodine-129 in seawater near the Fukushima Dai-ichi Nuclear Power Plant using a UV irradiation method

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Radioiodine ^{129}I is a critical radionuclide for radiation dose assessment in marine environments due to its accumulation in algae and its extremely long half-life (1.57×10^7 years). Since the environmental behavior and biological uptake of iodine depend on its chemical form, determining the concentrations of individual species is essential. However, data on organic ^{129}I remain limited because analysis requires large-scale experimental procedures, including UV irradiation, low-background sample preparation, and Accelerator Mass Spectrometry (AMS). Consequently, the behavior of organic ^{129}I in the marine environment remains poorly understood.

This study aims to investigate the concentrations and speciation of ^{129}I in seawater collected near the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) by applying a UV irradiation method originally reported by Otosaka et al. for organic ^{14}C [1]. The resulting data will contribute to a better understanding of ^{129}I behavior around the FDNPP and to improve the accuracy of radiation dose assessments in marine ecosystems.

In 2022–2025, seawater samples were collected near the FDNPP. Samples were processed at the LLRL, Kanazawa University, using solvent extraction methods. Extracted iodine was precipitated as AgI, and the $^{129}\text{I}/^{127}\text{I}$ ratio was measured via AMS at the University of Tsukuba [2], while ^{127}I concentrations were determined via Inductively Coupled Plasma Mass Spectrometry (ICP-MS). A mercury-lamp UV irradiation system was introduced for the pretreatment of organic iodine analysis (Figure 1), and preliminary measurements were conducted using seawater samples collected near the FDNPP.

^{129}I concentrations within a 2 km radius of the FDNPP during 2022–2025 ranged from 10^{-8} to 10^{-5} Bq/L, showing significant spatial variability. These results suggest that ongoing sources of ^{129}I within or near the FDNPP site have continued even prior to the ALPS-treated water discharge. Since the discharge began in August 2023, these spatial differences have become more pronounced, with elevated ^{129}I concentrations associated with the ALPS-treated water plume and potential leakage pathways. Simulated concentration levels generally matched observations, indicating that dispersion modeling reasonably reproduces ^{129}I distribution near the plant.

However, no clear increase in concentration was observed when comparing pre- and post-discharge data, suggesting that evaluating the impact of ALPS-treated water discharge based solely on monitoring is challenging. Averaged concentration data from numerical simulations may prove more effective for evaluating discharge-related increases. Furthermore, the persistent leakage of radionuclides from the site may exert a non-negligible influence compared to the controlled discharge, highlighting the need to identify the magnitude and sources of such leakage.

Preliminary UV irradiation experiments indicated an increase in inorganic ^{127}I concentrations, revealing

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an organic-to-inorganic iodine ratio of approximately 1:9. This result is consistent with reported iodine speciation in seawater, confirming the method's applicability to organic ^{127}I analysis. Conversely, no significant change was observed in inorganic ^{129}I concentration before and after UV irradiation, suggesting that the organic fraction of ^{129}I in these samples is relatively small. Future work will focus on further ^{129}I speciation analyses, integrated analysis with other isotopes (e.g., ^{137}Cs , ^3H), and the use of dynamic transfer models for marine organisms.

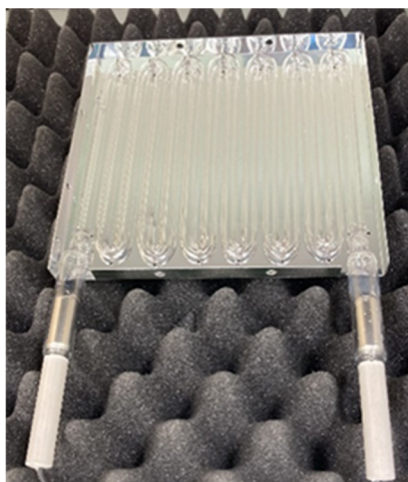


Fig. 1. Mercury-lamp UV irradiation system.
Seawater samples are placed in quartz vessels and irradiated with UV light for 24 hours to mineralize organic compounds into inorganic forms.

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4.

ION-BEAM IRRADIATION EFFECT

4.1 Raman scattering spectra of silica glass irradiated with 80 MeV I^{13+} ions

K. Awazu, K. Sasa, A. Uedono

For the realization of lunar bases and uncrewed lunar vehicles, long-term stable operation of advanced semiconductors, optoelectronic devices, and high-capacity optical fibers is crucial. The radiation environment on the lunar surface is significantly harsher than on Earth, with annual doses reported to be approximately 100–200 times higher. In the lunar radiation environment, protons and helium are the main components. Still, the heavy particle component, which is less significant as a flux, exhibits extremely high energy transfer and cannot be ignored from the perspective of material damage. In silica glass, high-density electron excitation and the resulting generation of delta electrons accompanying the passage of swift heavy particles induce nanoscale structural changes. This structural change causes abnormalities in electrical properties, dielectric breakdown, decreased light transmittance, and luminescence. Therefore, we analyzed the damage to silica glass induced by swift heavy particles using Raman spectroscopy [1].

The spatial resolution of micro-Raman scattering was 1 micron in the x and y directions and 2 microns in the depth direction. Fig.1 shows the Raman spectra of silica glass before and after irradiation with an 80 MeV I^{13+} beam at a fluence of $1 \times 10^{13} \text{ cm}^{-2}$, measured at the surface. The spectra before and after irradiation are shown in black and red, respectively. Before irradiation, the Boson peak is located below 50 cm^{-1} , and its position is unclear because the spectrometer can only detect signals above 50 cm^{-1} . After irradiation, it shifts to the higher wavenumber side, and a peak appears at 82 cm^{-1} . Furthermore, the Raman scattering intensity below 450 cm^{-1} decreases after irradiation, while the intensity of the Raman scattering at 609 cm^{-1} , which is attributed to 3MR, increases significantly. Since large-ring-size Raman scattering, such as 6MR and 8MR, has peaks below 450 cm^{-1} , this indicates that the large ring changed into a smaller ring, such as 3MR, after irradiation.

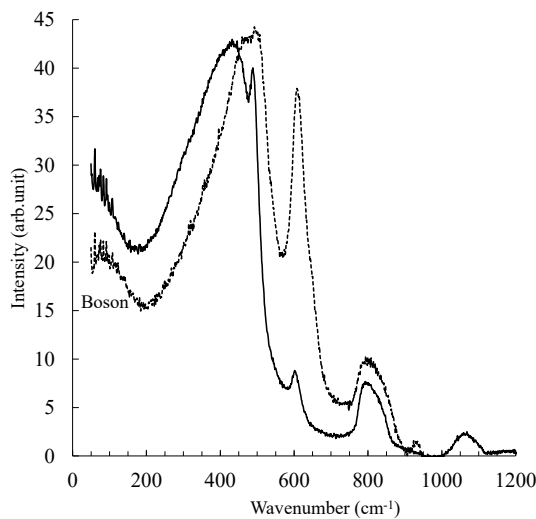


Fig. 1. Raman scattering spectra of silica glass irradiated with 80 MeV I^{13+} ion. Black line: pristine silica glass, red line: irradiated silica glass.

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4.2 Flux pinning effect by spherical irradiation defects arranged in series for high- T_c superconducting thin films and tapes

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Further improvement of in-field critical current density J_c in high- T_c superconductor REBa₂Cu₃O_y (REBCO, RE: Y or rare earth elements)-based coated conductors (CCs) is required, independent of magnetic field orientation, which leads to a reduction in the volume and cost of CCs for high magnetic field magnet applications [1], [2]. A fundamental approach for tuning the in-field J_c is to impede the motion of flux lines penetrating superconductors by introducing appropriate crystalline defects that act as pinning centers (PCs). Optimizing the morphology of PCs is one of the key factors for approaching a theoretical limit of J_c , i.e., a depairing current density J_d .

Columnar defects (CDs) with a linear shape act as strong PCs only when the magnetic field is oriented along their length [3]. By contrast, an isotropic pinning force against the magnetic field orientation is obtained for spherical defects (SDs) such as nano-particles, whereas the elementary pinning force of SDs is weaker than that of CDs [4]. When SDs are arranged in a series, they are expected to offer the advantages of both CDs and SDs: SDs arranged in a series can be expected to act as completely isotropic PCs for multiple flux lines due to their spherical shapes, and as intensive PCs for a single flux line through their alignment in a row. This morphology can be achieved by adjusting the electronic stopping power S_e for heavy-ion irradiation to high- T_c superconducting materials [5]. In this study, we investigated the flux pinning properties of REBCO films and CCs irradiated with a 44 MeV Au ion beam parallel to the c -axis, in order to systematically extract unique pinning features of SDs arranged in series.

The film samples used in this study were NdBCO films with a thickness of 400 nm, which were purchased from CERACO Ceramic Coating GmbH. We also used commercially available CCs with and without artificial PCs (APCs) from Faraday Factory Japan LLC (FFJ). The REBCO layers were prepared by a pulsed laser deposition process. The REBCO CCs with and without APCs were composed of a 2.9 μm -thick YBCO film including Y₂O₃ nano-particles and a 1.3 μm -thick GdBCO film, respectively. The Y₂O₃ particles with sizes of 5–20 nm are distributed at a density of $4000 \pm 2000 \mu\text{m}^{-2}$ in the YBCO CCs [4].

The irradiation with 44 MeV Au ions was performed using the 6 MV tandem accelerator at the University of Tsukuba, Japan. The ion beam was directed perpendicular to the film surface (i.e., parallel to the c -axis). The morphology of heavy-ion irradiation defects depends on the electronic stopping power S_e of the irradiated ions for the target material. In particular, irradiated ions with a very low S_e value of less than 12 keV/nm, such as 44 MeV Au ions, produce SDs aligned in a row along the ion path [5]. The fluences were in the range of 1.0×10^{11} to 5.0×10^{11} ions/cm², corresponding to equivalent matching fields of $B_\phi = 2$ T to 10 T.

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Fig. 1 shows a cross-sectional transmission electron microscopy (TEM) image of the irradiated YBCO CC with APCs, where the irradiation dose of 44 MeV Au ions corresponds to $B_\phi = 5$ T. The positions of SDs arranged in series are indicated by dashed arrows. The SDs, about 10 nm in size, are aligned in a row with gaps of 20–40 nm between them. Some of the SDs arranged in series are inclined at a slight angle to the c -axis. A similar irradiation defect morphology was observed in YBCO films irradiated using the same 44 MeV Au ions [5]. In addition, platelet-like nano-inclusions with sizes of 5–10 nm were also found in the irradiated YBCO CC with APCs (indicated by short arrows).

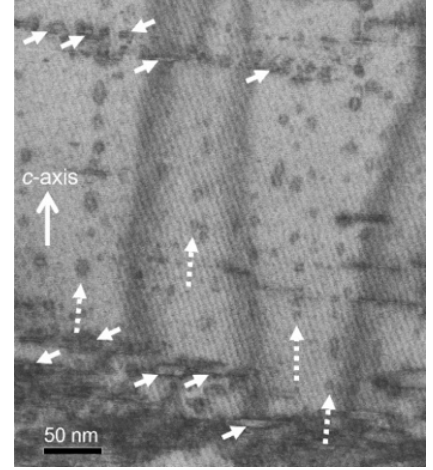


Fig. 1. Cross-sectional TEM image of a YBCO CC with APCs, which was irradiated with 44 MeV Au ions in the c -axis direction.

To get insight into the inherent nature of flux pinning by SDs arranged in series, we examined the thermal relaxation rate of magnetization. Fig. 2 shows the temperature dependence of normalized relaxation rate S . For all the samples, the S values show almost a plateau behavior around 0.02–0.03 for 20–60 K, which is a sign of the collective creep from random PCs [6]. Therefore, the introduced SDs act like random PCs, even though the SDs are aligned in a row. The introduction of correlated PCs such as CDs induces a peak of S around 25 K, which is attributed to flux creep through the double kink excitation of flux lines between neighboring CDs [7]. The peak of S almost disappears for the configuration of SDs arranged in series. However, a slight peak in S occurs around 20–25 K at the highest fluence of $B_\phi = 10$ T for all irradiated samples. When neighboring SDs arranged in series are close to each other, the flux pinning may exhibit a correlated nature: the double kink excitation can be generated between the neighboring SDs arranged in series in a quasi-correlated manner.

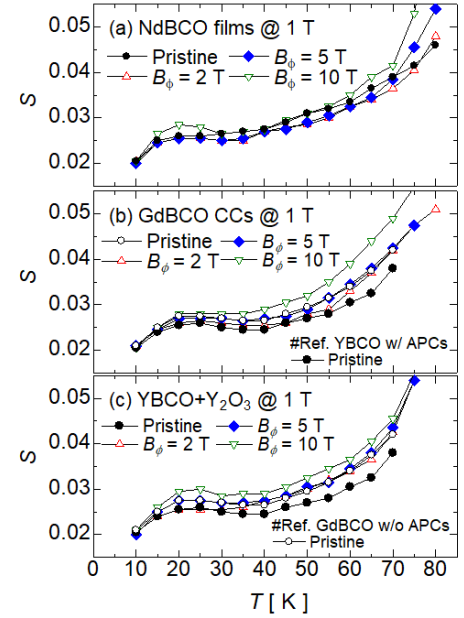


Fig. 2. Temperature dependence of normalized relaxation rate S as a function of irradiation fluence for (a) NdBCO films, (b) GdBCO CCs, and (c) YBCO CCs with APCs, respectively.

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4.3 Improvement of SEE tolerance in SiC trench power MOSFETs

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Silicon carbide (SiC) power devices are considered highly promising candidates for space applications due to their superior physical and electronic properties. However, several challenges have been identified when SiC devices are exposed to harsh radiation environments, including Single Event Effects (SEEs). Among these, Single Event Burnout (SEB) and Single Event Gate Rupture (SEGR) can lead to catastrophic device failure [1-3]. In this study, three types of SiC trench power MOSFETs were subjected to heavy-ion irradiation tests to evaluate and compare their susceptibility to SEEs.

Fig. 1 illustrates the cross-sectional structures of the three types of SiC MOSFETs fabricated at MIRISE Technologies Corporation and evaluated in this study. All devices share a common first drift n-layer. A distinguishing feature of MOSFETs (B) and (C) is the insertion of additional drift layers between the first drift and buffer layers; MOSFET (B) has two drift layers, while MOSFET (C) has three. A 60 MeV Al^{9+} beam from the 6 MV tandem accelerator at UTTAC was used for the irradiation tests. The device chips were irradiated in the direction perpendicular to their surfaces with a fluence set at 3.0×10^5 ions/cm². In this case, the LET and the range in SiC are 11.9 MeV/(mg/cm²) and 15.3 μm , respectively (Table 1). The range in SiC exceeds the total thickness of the drift layers. The drain current (I_D) and the gate current (I_G) were monitored during the irradiation.

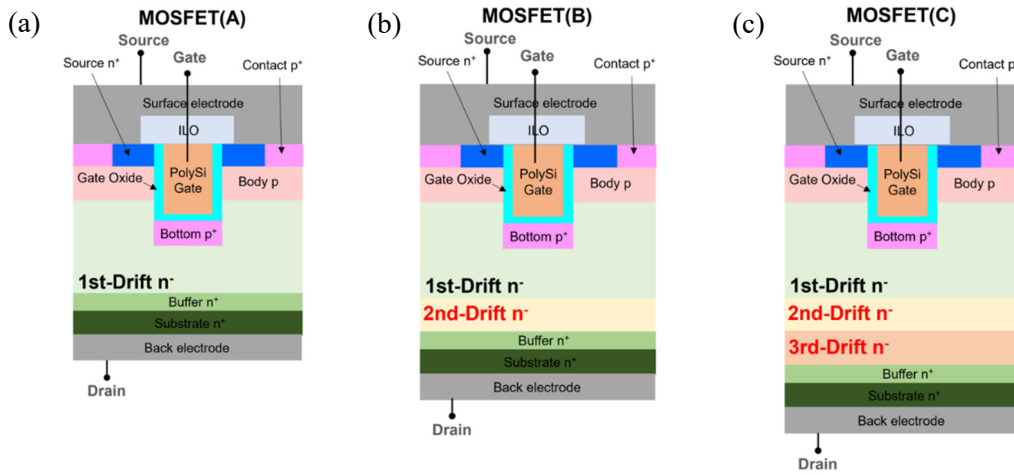


Fig. 1. Cross sections of the three types of SiC MOSFETs used in this study. (a) single drift layer, (b) double drift layers, and (c) triple drift layers.

Table 1. Characteristics of Al^{9+} ions incident on SiC, calculated using SRIM.

Ion species	Energy [MeV]	LET at the SiC surface [MeV/(mg/cm ²)]	Range in SiC [μm]
Al^{9+}	60	11.9	15.3

¹Japan Aerospace Exploration Agency (JAXA), M. Iwata was with JAXA at the time of this work

²MIRISE Technologies Corporation

Fig. 2 shows the transitions of I_D and I_G as functions of the applied drain voltage (V_D) during the irradiation at a V_G of -2 V. In MOSFET (A), a rapid increase in drain current was observed at V_D of 460 V, indicating the occurrence of SEE. In MOSFET (B) with double drift layers, a rapid increase in drain current was observed at V_D of 520 V. This means that the SEE tolerance improved by 60 V. Moreover, in MOSFET (C) with triple drift layers, a rapid increase in drain current was observed at V_D of 600 V, indicating that the SEE tolerance improved by 140 V. These findings show that differences in the number of drift layers affect SEE tolerance, and the triple-drift-layer structure further improves SEE tolerance. This suggests that the multiple drift layers contribute to electric field relaxation within the device, thereby improving SEE tolerance.

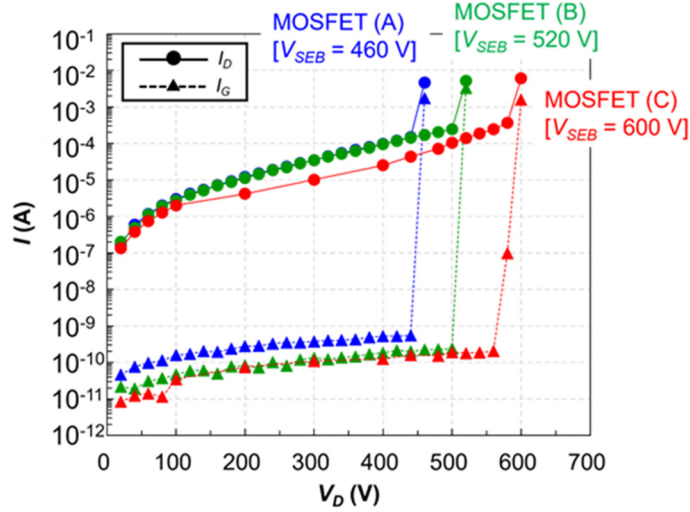


Fig. 2. Results of I_D and I_G transitions during the Al^{9+} irradiation of MOSFETs (A), (B), and (C).

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4.4 Single-Event Burnout Evaluation of HEMT Devices for Millimeter Wave Wireless Power Transmission

A. K. Sugihara¹, M. Irie², A. Wakejima²

The authors are developing high-frequency, high-efficiency and high-power HEMT devices, based on a novel GaN-on-GaN process, for lunar Wireless Power Transmission (WPT) applications. On the lunar surface, heavy particles of solar and galactic origins play the dominant role in defining the radiation environment. Consequently, survivability of single events is crucial for lunar applications.

In this work, we evaluate the condition at which Single-Event Burnout (SEB) may occur on the prototyped HEMT devices. The evaluation is a destructive test, based on the following procedure, typically followed when evaluating SEBs on power semiconductors. Device Under Test (DUT) is irradiated with ions with sufficient energy to reach the DUT's vulnerable area. The semiconductor is then biased with gradually increasing bias voltage, until SEBs can be observed.

The setup and results are summarized in Fig. 1. Ag^{9+} ions are accelerated to 55 MeV, and irradiated on the DUT with fluence of 30000–60000 p/cm²/s. The DUT is biased from 10 V with 10 V steps to 60 V. The above fluence is maintained for approximately 120 seconds before the bias is increased to the next step.

The results are tabulated (Fig. 1), showing that the majority of the prototyped devices survived to a bias of 60 V, which is close to the prototype's breakdown voltage. This result indicates the effectiveness of the new GaN-on-GaN process against SEBs.

The DUT's high-frequency characteristics have been measured separately with promising results. Future work will focus on increasing the device's breakdown voltage, which should directly translate to improved resilience against SEB in lunar environment.



		SEB Voltage [V]	Fluence [p/cm ² /s]	Breakdown Mode
DUT 2 -1	FET13	50	68000	Drain short
	FET15	40	68000	Drain short
	FET19	30	68000	
	FET20			Drain short
DUT2-3	FET8	20	60000	Drain short
	FET16	30	60000	Drain short
	FET18	50	60000	Drain short
	FET19	60	60000	Drain short
	FET20	20	60000	Drain short
DUT3- 2	FET3	60	32000	Drain short
	FET16	60	32000	Gate,Drain short
	FET17	65	32000	Gate,Drain short
	FET21	60	64000	Drain short
	FET22	60	64000	Drain short

Fig. 1. DUT setup in the 6MV Tandem Accelerator (top) and the recorded bias voltage at SEBs (bottom)

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²Kumamoto University

4.5 Surface resistance control of sapphire substrates by Pt ion implantation

A. Ogura, T. Kaneko, S. Ishii, S. Katagiri

Solid insulators are indispensable for supporting accelerating electrodes in charged-particle accelerators operated under high vacuum and high voltage, such as electron microscopes and X-ray CT systems. Surface flashover occurring on the insulator surface exhibits a lower withstand voltage than vacuum gap breakdown, which has become an obstacle to further increases in operating voltage and device miniaturization. One of the causes of this flashover is positive charging on the insulator surface. As a method for suppressing this positive charging, techniques that provide slight surface conductivity to the insulator have been investigated, including coating and ion implantation [1, 2]. We have previously demonstrated that the surface resistance of sapphire substrates can be controlled by irradiating them with 300 keV Ni ions [3]. However, because the ions did not accumulate near the surface under high-energy irradiation, we attempted to control the ion distribution so that ions accumulated at the topmost surface of sapphire by using Ti film coatings and reactive dry etching. In addition, since the irradiation area was limited to only a few millimeters, it was difficult to investigate the charging and discharge characteristics. Therefore, in this study, we investigated low-energy ion implantation to achieve ion accumulation near the topmost surface, as well as implantation methods for fabricating large-area samples.

The ion implantation experiments were carried out using the 9F ion source operated by the University of Tsukuba Tandem Accelerator Complex (UTTAC). As ion species, Si, Ni, and Pt were investigated. For Si and Ni implantation, numerical estimations indicated that, in order to accumulate ions within the topmost ~20 nm region, the acceleration energy had to be reduced to 20 keV and the samples had to be irradiated at an incident angle greater than 50°. Because the beam current becomes significantly lower at 20 keV, Ni implantation was instead performed at 30 keV. Under these conditions, the ion beam current was approximately 250 nA for Si at 20 keV and 29 nA for Ni at 30 keV. Based on the previous year's results, it is known that ion fluences on the order of 10^{16} cm^{-2} are required to control the surface resistance. Therefore, for large-area irradiation, a beam current of at least 1 μA is necessary. From this viewpoint, Si and Ni were judged to be unsuitable as ion species. The ion beam current decreases significantly at lower acceleration energies due to the characteristics of the ion source. Therefore, it is desirable to use as high an acceleration energy as possible while still enabling ion accumulation near the topmost surface. For this reason, Pt, a heavy ion species, was investigated. Numerical calculations showed that irradiation of a sample tilted by 50° with 100 keV Pt ions would result in ion accumulation within approximately 35 nm from the surface. In addition, a beam current of around 1 μA could be secured, making Pt ions suitable for implantation. Furthermore, based on the previous year's Ni ion implantation results, it was found that the formation of nanoclusters by the implanted ions inside sapphire is important. Since Pt is a highly stable metal, it is expected to be hardly oxidized and to readily form nanoclusters.

Figure 1 shows the fluence dependence of the surface resistance of sapphire. It was confirmed that the surface resistance could be controlled at a fluence around 10^{16} cm^{-2} . In addition, the data exhibit a linear relationship on a log-log plot, indicating a power-law dependence of the surface resistance on ion fluence.

Figure 2 shows a HAADF-STEM image of the sample irradiated with a fluence of 10^{16} cm^{-2} . Pt nanoclusters with sizes of several nanometers can be observed accumulating within approximately 34 nm from the topmost surface. Similar to the case of Ni implantation, it is expected that electrical conduction occurs through tunneling between neighboring nanoclusters. Figure 3 shows beam scanning achieved using the electrostatic deflector newly installed in the accelerator system. It can be confirmed that the ion beam was deflected in both the X and Y directions by a bipolar power supply. In future work, large-area samples will be fabricated using this beam-scanning technique, and their surface charging behavior and flashover withstand voltage will be investigated.

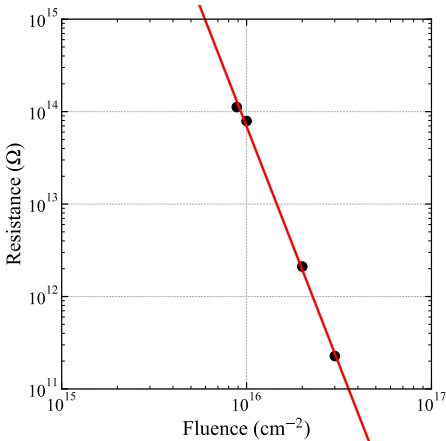


Fig. 1. Surface resistance of sapphire as a function of ion fluence.

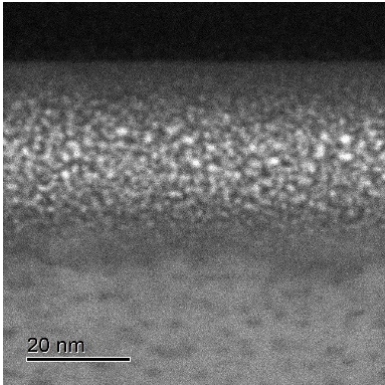


Fig. 2. Cross-sectional HAADF-STEM image of sapphire irradiated with $1 \times 10^{16} \text{ cm}^{-2}$ Pt ions.

Ch1→ ↓Ch2	-3 kV	0 kV	3 kV
-3 kV			
0 kV			
3 kV			

Fig. 3. Ion beam scanning pattern obtained using a bipolar power supply.

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5.

MATERIALS RESEARCH

5.1 Characterization of plasma-irradiated SiO₂ films deposited on Si using positron annihilation spectroscopy

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The increase in interconnect density due to transistor scaling has become a severe bottleneck for modern high-performance computing. To overcome this issue, 3D integration technology has emerged. A core technology of 3D stacking is wafer-to-wafer bonding. In the conventional bonding process, the SiO₂ surface is activated by plasma treatment. In the present study, we used a monoenergetic positron beam to probe defects introduced by plasma activation in SiO₂ layers deposited on Si substrates.

The sample structure used in the present experiments was SiO₂/Si. The SiO₂ layers were deposited on Si wafers by plasma-enhanced chemical vapor deposition using liquid-based tetraethyl orthosilicate (TEOS) as the precursor. The radio-frequency power, pressure, and substrate temperature during deposition were 1 kW, 80 Pa, and 300 °C. Defects introduced by the plasma activation were probed by using a monoenergetic positron beam. This technique is described in detail elsewhere [1].

Figure 1 shows the S values of the SiO₂/Si samples before and after plasma activation as a function of incident positron energy E . Before the measurements, the samples were stored in an air atmosphere at room temperature. The S values were measured one day [N₂(1st)] and one week [N₂(2nd)] after the process. For the sample before plasma activation (as-received), positrons implanted at $E < 10$ keV mainly annihilated in the SiO₂ layer. For the samples before and after the plasma activation, the solid curves fit the experimental data. The obtained depth distributions of S are shown in the inset of Fig. 1. For the samples after N₂-plasma activation, S near the surface ($E < 4$ keV) decreased. The primary cause of the decrease in S is positron/electron traps, such as dangling bonds, which are considered to be introduced by electron/ultraviolet irradiation from plasma, as well as the physical impact of molecules [1]. For the sample characterized one week after the plasma activation, S in the subsurface region increased but was still smaller than S for the sample before the activation. The observed recovery of S can be attributed to the adsorption of organic molecules containing carbon and nitrogen, which decreases the concentration of positron/electron traps by terminating dangling bonds. The present research showed that positron annihilation can provide information on defects introduced by plasma treatment in the subsurface region of SiO₂.

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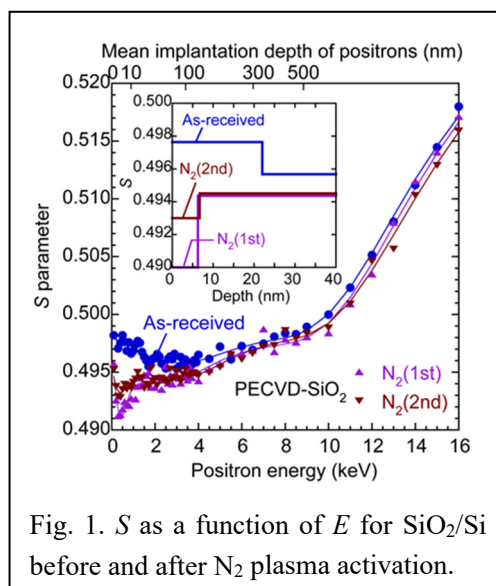


Fig. 1. S as a function of E for SiO₂/Si before and after N₂ plasma activation.

¹Yokohama National University

²Toray Engineering Co., Ltd.

³Tohoku University

5.2 Vicinage effects on convoy electron yield from carbon foil under LiH^+ bombardment

Y. Yano, T. Noguchi, T. Manako, K. Ishimoto, S. Ishii, K. Sasa, S. Tomita

Convoy electrons are forward-emitted electrons with velocities matching those of emerging projectiles from solid targets, first observed as a characteristic cusp-shaped peak in the forward electron-emission spectrum [1]. They are understood to form through the buildup of a closely phase-space-correlated electron flow in the proximity of the projectile ion inside the solid, effectively being dragged along by its Coulomb field [2]. Previous investigations using C_n^+ molecular ions demonstrated a substantial enhancement in convoy electron yield compared to the sum of those of the constituent atomic ions [3,4]. This enhancement is known as the vicinage effect [5] and arises from the short distances between the constituent atomic ions dissociated from the molecular ions in the solid target.

While previous studies on the vicinage effect have focused primarily on homonuclear molecular ions, investigations involving heteronuclear molecular ions remain limited. In heteronuclear molecular ions, the stopping powers of the constituent atoms differ, causing the relative velocity between the constituent ions to increase with target thickness. We previously confirmed that irradiation of a carbon foil ($4.7 \mu\text{g}/\text{cm}^2$) with the heteronuclear molecular ion OH^+ also produces an enhancement of the convoy electron yield due to the vicinage effect, as observed for homonuclear molecular ions [6]. However, at this thickness, the velocity difference between the O and H ions was small, and it was not possible to investigate the vicinage effect in detail under conditions of sufficiently large relative velocity. In the present study, we therefore irradiated a thicker carbon foil with the heteronuclear molecular ion LiH^+ to investigate the vicinage effect under conditions of larger relative velocity.

Experiments were conducted using the 1 MV Tandetron accelerator at UTTAC. LiH^+ , Li^+ , and H^+ ions were accelerated to 120 keV/u and directed onto carbon foil targets of $44.4 \mu\text{g}/\text{cm}^2$. Since this accelerator selects ions by mass-to-charge ratio (m/q), contamination of ${}^6\text{LiH}_2^+$ ($m/q = 8$) in the ${}^7\text{LiH}^+$ beam cannot be excluded in ordinary operations. To avoid this, ${}^7\text{LiH}^+$ was selected using a sector magnet after acceleration, by separating it from the primary beam of ${}^7\text{LiH}_2^+$ ($m/q = 9$) from the ion source. Convoy electrons were measured using an electrostatic energy analyzer positioned at 0° relative to the direction of the beam. Energy scans were performed, with data acquisition normalized to the integrated ion current at each energy step to compensate for beam-intensity variations. The correspondence between the ion current and the number of incident particles was carefully determined in a separate experiment.

Figure 1 shows the electron energy spectra obtained for LiH^+ , Li^+ , and H^+ ions incident on a carbon foil of $44.4 \mu\text{g}/\text{cm}^2$. The electron yields are normalized to the number of incident particles. As seen in the spectra, peaks are observed around 55 eV for LiH^+ and Li^+ irradiation, while a peak appears around 50 eV for H^+ irradiation. These peaks correspond to convoy electrons. Positions of vertical lines represent the velocities of Li (red) and H (blue) ions after passing through the $44.4 \mu\text{g}/\text{cm}^2$ carbon foil, as estimated with SRIM [7].

As seen in Fig. 1, the convoy electron peak of $Y_e(\text{LiH}^+)$ is larger than the sum of $Y_e(\text{Li}^+)$ and $Y_e(\text{H}^+)$, indicating an enhancement of the convoy electron yield due to the vicinage effect in the $44.4 \mu\text{g}/\text{cm}^2$ carbon

foil, consistent with the previous result for OH^+ [6]. Furthermore, the peak of $Y_e(\text{LiH}^+) - (Y_e(\text{Li}^+) + Y_e(\text{H}^+))$ lies between those of $Y_e(\text{Li}^+)$ and $Y_e(\text{H}^+)$. This suggests that, in addition to the convoy electrons dragged along by the Coulomb field of each constituent ion, there exist convoy electrons that are simultaneously subject to the Coulomb fields of both the Li and H ions, and it is this additional population that manifests as the observed yield enhancement. If the stopping power acts on the Li and H ions in the same manner as for monatomic ions, the inter-ion distance between Li and H is estimated to be at least 57.4 Å due solely to the spread arising from the difference in stopping power. Moreover, if the Coulomb explosion spread, estimated from the equilibrium charges of Li and H ions at 120 keV/u, is added to this stopping-power-induced spread, the inter-ion distance reaches as large as 75.8 Å.

These results suggest that, despite the velocity difference between the Li and H ions dissociated from LiH^+ within the solid, the convoy electrons can be influenced by the Coulomb potentials of both ions. This implies that convoy electrons are not exclusively associated with a single constituent ion, but are instead subject to the collective Coulomb influence of multiple ions during transmission. Given that the inter-ion separation estimated reaches several tens of angstroms, these findings suggest that the vicinage effect can operate even when the constituent ions are separated by distances on the order of 50–100 Å.

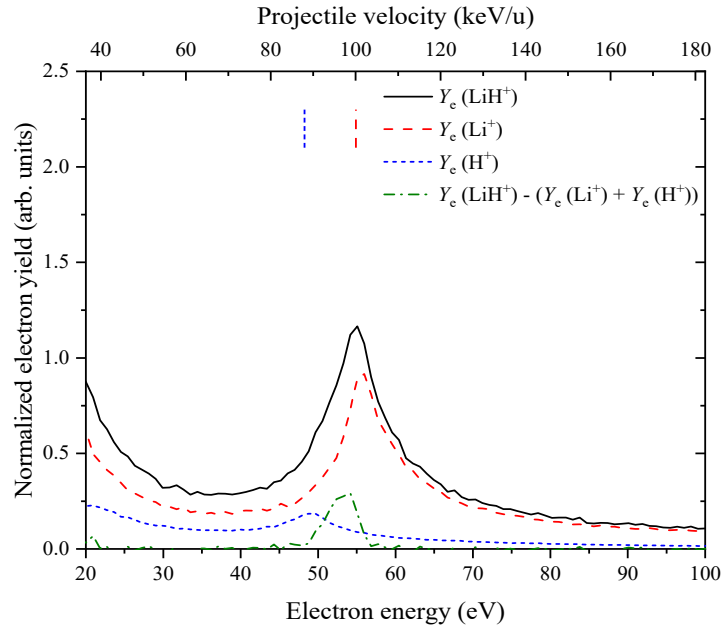


Fig. 1. Electron energy spectra obtained for a $44.4 \mu\text{g}/\text{cm}^2$ carbon foil under 120 keV/u LiH^+ (black line), Li^+ (red dashed line), and H^+ (blue dashed line) irradiation, along with the difference between the LiH^+ spectrum and the sum of the Li^+ and H^+ spectra (green dotted line).

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5.3 An investigation of an integrated proton-dynamic therapy, iPDT, using the nuclear resonance reaction between the stable nitrogen isotope ^{15}N and a proton beam

H. Matsui, H. Kurokawa, A. Taninaka, I. Suzuki, Y. Kobayashi, K. Sasa, S. Ishii

This study aimed to evaluate the efficacy and safety of integrated proton-dynamic therapy (iPDT) using ^{15}N -labeled 5-aminolevulinic acid (^{15}N 5-ALA) through both in vitro and in vivo investigations. In the co-culture system of normal rat gastric epithelial cells (RGM) and transformed gastric cancer-like cells (RGK), proton irradiation was performed after administration of either conventional 5-ALA or ^{15}N 5-ALA. Forty-eight hours after irradiation, the area occupied by RGK cells was significantly suppressed in the ^{15}N 5-ALA-treated group compared with proton irradiation alone, suggesting a tumor-selective inhibitory effect.

Immunofluorescence analyses demonstrated enhanced DNA damage responses in the ^{15}N 5-ALA-treated group. γH2AX expression was markedly increased at 120 minutes after irradiation, while 53BP1 expression remained elevated throughout the observation period, supporting the existence of additional DNA damage induced by the nuclear resonance reaction. Confocal microscopy further revealed characteristic nuclear morphological alterations in cancer cells. Similar enhancement of γH2AX expression was confirmed in U87 glioma cells by Western blotting analysis.

In vivo safety evaluation using mice demonstrated no significant differences in blood biochemical parameters, including Cre, BUN, ALT, and AST, between the ^{15}N 5-ALA-treated and control groups, indicating favorable safety profiles across the tested dosage range. Experiments using tumor-bearing mouse tissue sections are currently ongoing to obtain preliminary therapeutic efficacy data.

In addition, preparations for a clinical protocol to evaluate tumor-specific accumulation of ^{15}N 5-ALA are in progress. These findings suggest that iPDT combined with ^{15}N 5-ALA represents a promising strategy for enhancing tumor-selective proton beam therapy.

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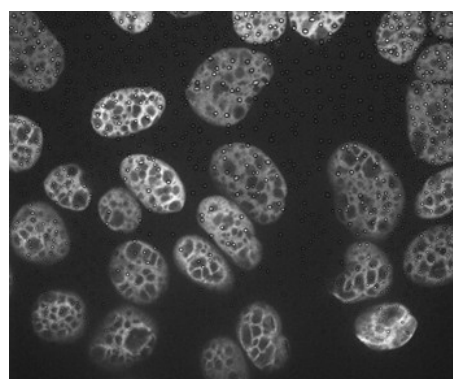


Fig. 1. **Representative image of CND:** Cobblestone-like defective structures observed within nuclei after resonant nuclear reaction. The relationship between these structures and existing modes of cell death will be investigated.

5.4 Magnetic property of outer oxides layer on metallic ferromagnetic particles

E. Kita, H. Yanagihara

The magnetic recording media has been used to record sound and music, additionally video recording and mass data storage systems. Particulate magnetic media have been useful materials with high performance and low cost. Due to the demand on high density recording, materials have been shifted from oxide to metallic materials because of high magnetic moments[1]. Oxide materials possess high chemical stability, while metallic ferromagnetic materials excel in physical properties but are often chemically unstable. Chemical stability has been improved by applying an anti-oxidation surface treatment to the surface of metallic materials. Such surface layers have amorphous structures and are considered superparamagnetic[2,3]. To understand the surface oxidization, Mössbauer spectroscopy was performed.

In recording tapes for an 8 mm video system, usually acicular metallic Fe particles were used as recording media. A commercial metal tape product of the Maxell company was used as a sample. The tape with a width of 8 mm was cut into pieces 12 mm in length, and the sample was prepared by piling up six layers of the pieces. Mössbauer spectra were gathered over the sample temperature range between 3 K and room temperature.

Figure 1 shows the temperature dependence of Mössbauer spectra of the magnetic recording tape. The spectrum at room temperature can be fitted with a combination of a ferromagnetic sextet originating from metallic bcc iron and a doublet. Additionally, there is a sextet with a small area, likely originating from stable iron oxide. At 3 K, the doublet completely disappeared and a broadened sextet with large hyperfine field appeared outside of metallic Fe. It is concluded that a paramagnetic part at room temperature was magnetically ordered at 3 K.

The spectrum at 200 K resembled that at room temperature and was also fitted in the same manner as that at room temperature and the area ratios did not change much. The spectrum recorded at 100 K showed metallic Fe and the doublet seen at high temperature changed to a

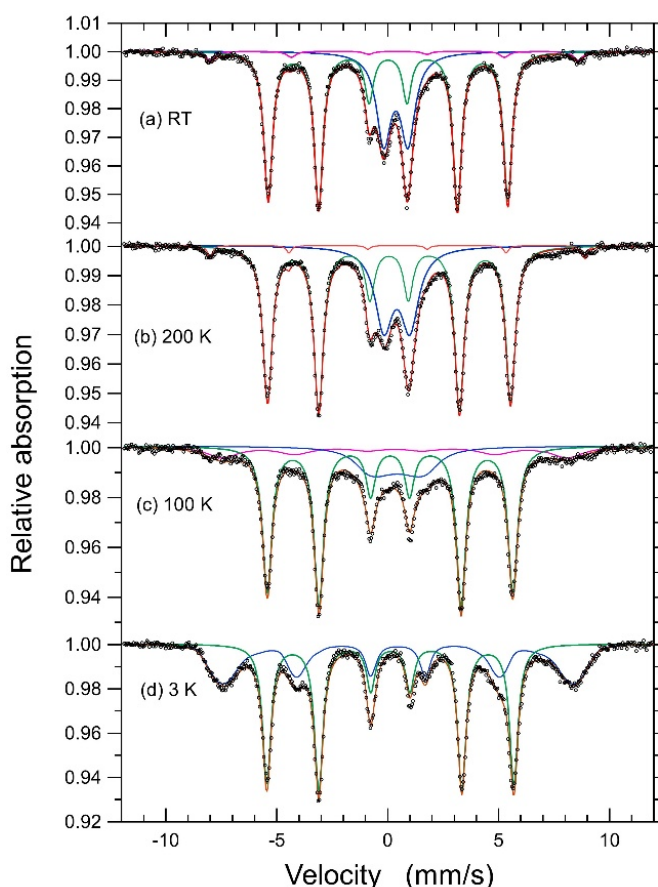


Fig. 1. Temperature variation of Mössbauer spectra for a metal tape. Spectra recorded at (a) room temperature, (b) 200 K, (c) 100 K and (d) 3 K are plotted.

broadened magnetic sextet with small internal field.

The area ratio of metallic Fe, corresponding to Mag 1 in Table 1, slightly decreased from 67 % at room temperature to 62 % at 3 K. The outer layer of metallic Fe particles has been reported to be oxidized and the ratio of them to the total Fe amount changed from 28 % to 38 %. The reason might be due to the temperature dependence of the recoil-free ratio related to the Debye temperature of the outermost layer. The recoil-free ratio can be expressed by the following equation based on the Debye model[4]. From the equation, the Debye temperature of the surface must be lower than that of the inner materials. Numerical estimation will be carried out for further discussion.

Table 1. Fitting Parameters of Mössbauer spectra of the magnetic recording tape

Temperature	Sub-spec	Hhf (kOe)	IS* (mm/s)	QS (mm/s)	Line Width (mm/s)	Area (%)
RT	Mag 1	335	0.016	-	0.466	67.8
	Mag 2	515	0.370	-0.170	0.513	3.9
	Doublet 1		0.366	1.089	0.786	28.3
200K	Mag 1	340	0.063		0.505	69.4
	Mag 2	525	0.442		0.280	2.3
	Doublet 1		0.412	1.17	0.972	28.3
100K	Mag 1	344	0.106		0.509	67.0
	Mag 2	483	0.317		1.779	14.8
	Mag 3	83	0.426		1.466	18.1
3 K	Mag 1	346	0.119		0.463	61.5
	Mag 2	479	0.465		0.606	38.5

* IS, isomer shift referred to room temperature natural Fe.

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5.5 Low temperature Mössbauer spectroscopy of Wüstite FeO

E. Kita, H. Yanagihara

FeO, MnO, CoO, and NiO have a rock-salt-type structure and have long been studied as typical materials in which superexchange interactions play an important role. A contribution of orbital angular momentum to the physical properties is expected in FeO. Because it is difficult to synthesize stoichiometric FeO, the magnetic structure in the low-temperature phase has not been definitively established. The transition temperature of FeO is reported to be around 190 K and depends on the oxygen content.[1] In high-pressure-synthesized samples[2], Mössbauer spectroscopy (MS) yields an almost singlet signal at room temperature, suggesting that the sample is homogeneous. In this study, low-temperature MS measurements were performed to investigate the magnetic transition and to consider the magnetic structure at low temperatures.

For sample preparation, α -Fe and α -Fe₂O₃ were mixed at a molar ratio corresponding to Fe:O = 1:1, and subjected to high-pressure synthesis using the belt-type apparatus at NIMS.[2] MS measurements were carried out between 3 K and room temperature using a cryostat. The lattice constant obtained from XRD was 4.316 Å.

Figure 1 shows the temperature dependence of the MS spectra. At room temperature, the spectrum exhibits an almost pure singlet, indicating a paramagnetic state down to 200 K. The paramagnetic component begins to decrease around 190 K, while the magnetically ordered phase gradually increases. At 180 K, an internal magnetic field of 264 kOe is observed. During this temperature range, the internal field of the magnetically ordered phase does not change significantly; instead, its spectral intensity increases as the system transitions from the paramagnetic to the magnetically ordered state. The abrupt change in internal field between 185 and 190 K cannot be explained by a second-order phase transition and is therefore identified as a first-order transition. The transition appears to be distributed over a few kelvin, suggesting an influence of deviation from the stoichiometric composition ($x = 1$). Meanwhile, the spectrum at 80 K differs from that at 180 K, implying an additional change in magnetic structure at low temperatures.

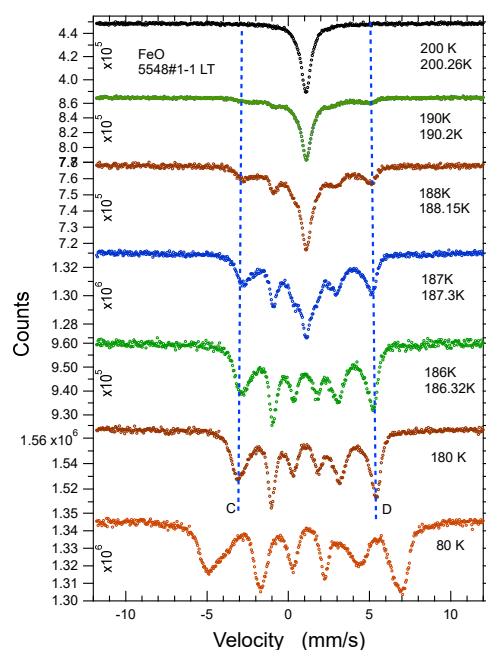


Fig. 1. Mössbauer spectra of FeO powder at temperatures between 80 and 200 K.

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5.6 Mössbauer study of Fe oxide thin films grown on Magnesium Aluminum Oxide (MAO) single crystal substrates

R. Waseda, H. Yanagihara and E. Kita

It is known that Fe, a ferromagnetic metal, exhibits perpendicular magnetic anisotropy (PMA) at interfaces with rock-salt-type metal oxides. Ferromagnetic Fe grown on MgO (Fe/MgO), a representative rock-salt oxide, shows strong PMA and has been applied in the field of spintronics. Fe also exhibits PMA on other rock-salt oxides such as NiO and CoO. Although FeO is likewise a rock-salt oxide, it is not as stable as NiO or CoO. In particular, the lattice constant is considered to have a significant influence on its physical properties. Therefore, to explore a method for growing FeO, we selected a spinel-type magnesium aluminum oxide (MAO, MgAl_2O_4) as the substrate and investigated whether FeO growth is feasible. Table 1 lists the lattice constants of the relevant materials.

Table 1. Lattice parameters and structure of related materials.

Material	α -Fe	FeO	MgO	NiO	Magnetite	MAO, MgAl_2O_4
Crystal Structure	bcc	NaCl	NaCl	NaCl	Spinel	Spinel
Lattice constant (nm)	0.287	0.428	0.422	0.418	0.840 (0.420)	0.8155 (0.408)

Taking FeO as the reference, all other oxides have smaller lattice constants. The lattice mismatch with MAO reaches 5%. Thin-film growth of Fe oxides was performed on single-crystal MAO substrates by reactive sputtering, introducing oxygen as a reactive gas into Ar gas. The obtained films were examined using Conversion Electron Mössbauer Spectroscopy (CEMS).

Figure 1 shows the Mössbauer spectra of samples grown under different oxygen flow rates. At an oxygen flow rate of 0.35 sccm, spectra from a small amount of α -Fe were observed in addition to the main FeO phase. At 0.40 sccm and 0.45 sccm, magnetite was detected as a secondary phase in addition to the main phase. These results indicate that the oxygen flow rate required to obtain single-phase FeO lies between 0.35 and 0.40 sccm. Compared with growth on MgO substrates, the growth window for FeO with respect to oxygen flow rate is narrower.

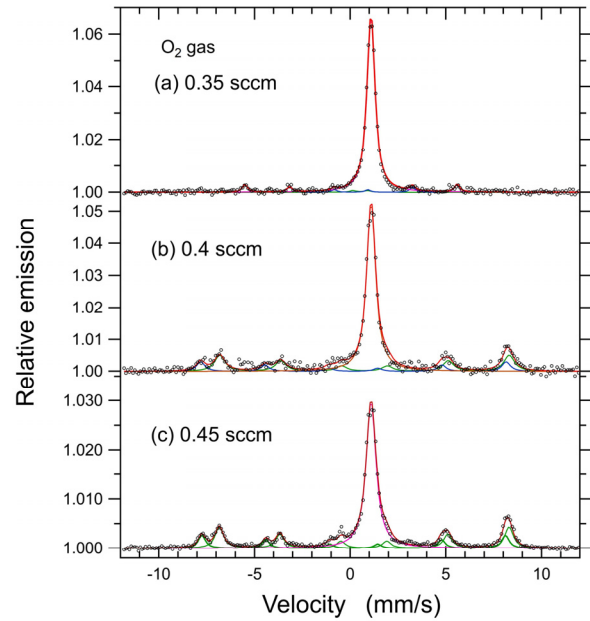


Fig. 1. Mössbauer spectra of FeO sputtered films grown with oxygen gas flow of (a) 0.35 sccm, (b) 0.4 sccm and (c) 0.45 sccm.

5.7 Low-temperature annealing-induced crystallization in an iron based amorphous alloy

R. Onodera¹, K. Takahashi², E. Kita

An iron-based nanocrystalline alloy known as NANOMET[1] is obtained by rapid heat treatment of an amorphous precursor at a heating rate of 400 K/min. This alloy has attracted considerable attention as a core material for high-power inductors because it exhibits excellent soft magnetic properties together with a high saturation magnetic flux density, B_s . In this study, heat-treatment processes were investigated to relax the stringent crystallization conditions required for the formation of NANOMET nanocrystalline structures. Several heat treatments were carried out at sufficiently low temperatures to avoid crystallization, as summarized in Table 1.

Heat treatments were performed using a cryogen-free superconducting magnet and an electric furnace at the High Field Laboratory for Superconducting Materials, Tohoku University. Mössbauer spectroscopy was performed at room temperature for sample pieces cut into 10×10 mm from the ribbon. Conventional constant acceleration mode was used for measurements with a velocity range of ± 12 mm/s. The velocity scale and isomer shift were calibrated via α -Fe at room temperature.

As shown in Figure 1, no significant changes in the Mössbauer spectrum are observed even after low-temperature heat treatment. In the spectral fitting, we assumed a distribution of the hyperfine field, HF. The average value of the HF obtained from the fitting increased slightly with heat treatment and increased further upon application of an external magnetic field during heat treatment (see Table 2).

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Table 1. Heat treatment conditions

Applied field	Annealing temperature	Condition
0 T	$0.85T_x$	(a)
	$0.90T_x$	(b)
10 T	$0.85T_x$	(c)
	$0.90T_x$	(d)
as-Q		(e)

T_x : the crystallization temperature at 10 K/min.

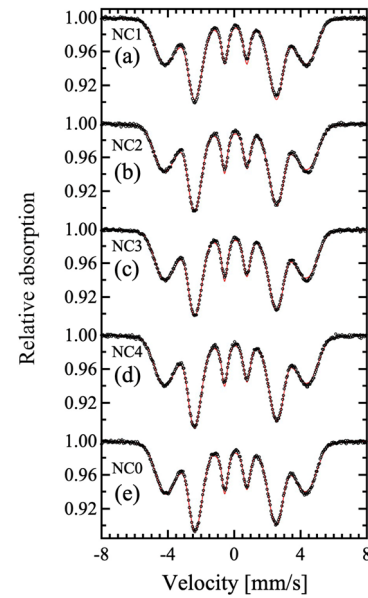


Fig. 1. Mössbauer spectra of sample ribbons after heat treatment (a) - (d), and of the as-Q ribbon (e).

Table 2. Heat treatment conditions

Heat treatment	Hyperfine field [T]
(a)	25.62
(b)	25.87
(c)	25.67
(d)	25.89
(e)	25.38

Hyperfine field of α -Fe: 33 T

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5.8 Evaluation of valence states in iron oxide nanoparticles for hyperthermia applications

H. Mamiya¹

It has been reported that the performance of iron oxide nanoparticles used for cancer hyperthermia therapy strongly depends on their valence state; however, the details remain unclear. In this study, we prepared two types of iron oxide nanoparticles with particle sizes of 17 and 19 nm for hyperthermia applications and evaluated their valence states using Mössbauer spectroscopy. Figure 1 shows the Mössbauer spectra and isomer shift values obtained from the analysis for these nanoparticles at high (200 K) and low (3 K) temperatures. The results indicate that the isomer shifts are similar to those of γ - Fe_2O_3 , and no splitting is observed on the low-velocity side of the magnetic sextet—a feature characteristic of Fe_3O_4 . These findings suggest that the nanoparticles are mainly composed of ferric (Fe^{3+}) iron.

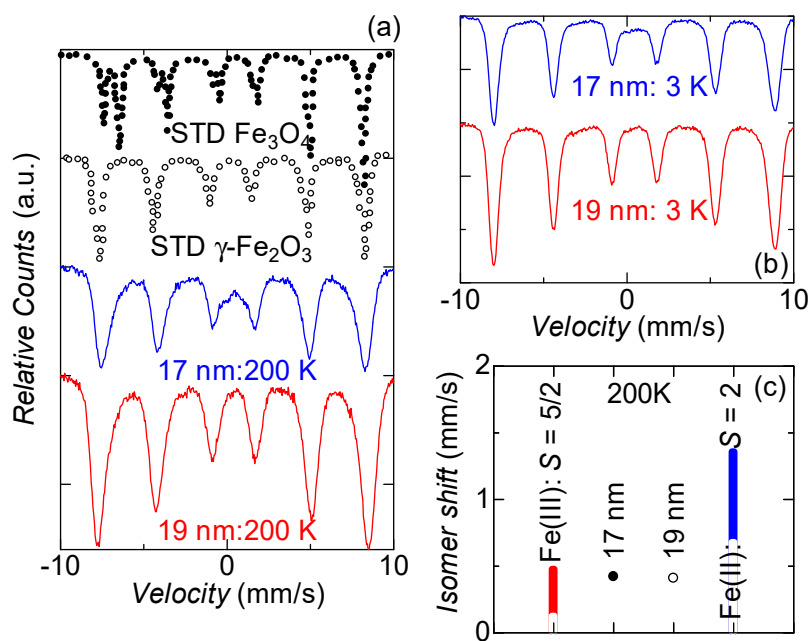


Fig. 1. Mössbauer spectra for nanoparticles in samples MF17 and MF19 at 200 K (a) and 3 K (b), as well as standard magnetite (Fe_3O_4) and maghemite ($\gamma\text{-Fe}_2\text{O}_3$) [1]. (c) shows the estimated values of the isomer shift at 200 K for comparison with Fe_3O_4 and $\gamma\text{-Fe}_2\text{O}_3$.

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71. Z. Shi, K. Sasa, M. Matsumura, T. Takahashi, Y. Takaku, M. Kuwae, S. Yamasaki, K. Sueki, A. Sakaguchi, “Isotopic Composition of ^{129}I in Marine Sediments from Beppu Bay: Assessing Its Feasibility as an Anthropocene Marker”, 第 26 回 AMS シンポジウム (JAMS-26), 名古屋大学 坂田・平田ホール, 名古屋市 (2026 年 3 月 26–27 日).

72. Z. Shi, Y. Takaku, S. Yamasaki, Y. Shigekawa, K. Sasa, M. Matsumura, K. Sueki, A. Sakaguchi, “Differential uptake of iodine species by the coral *Galaxea fascicularis* under controlled laboratory conditions”, 第 26 回 AMS シンポジウム (JAMS-26), 名古屋大学 坂田・平田ホール, 名古屋市 (2026 年 3 月 26–27 日).
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7.

THESES

Master theses

YANO, Yuichiro	Vicinage Effects on Convoy Electron Yield under Diatomic Molecular Ion Irradiation (二原子分子イオン照射におけるコンボイ電子収量の近接効果)
MATSUDA, Kazuma	Development of Particle Detectors Based on Superconducting Tunnel Junctions (超伝導トンネル接合を用いた粒子検出器の研究)
TAKEO, Kouki	Reduction of antiphase boundary defects in spinel ferrite epitaxial films via topotactic oxidation (スピネルフェライト薄膜中に生じた逆位相欠陥のポスト酸化による低減)
KAWABATA, Ryosuke (Tohoku University)	Study on Cs ₂ Li(Gd,Ce)Cl ₆ and BaCl ₂ Scintillators for a Novel Radiation Particle Identification Method Utilizing Emission Wavelength Information (発光波長情報を用いた新規放射線粒子識別法のための Cs ₂ Li(Gd,Ce)Cl ₆ 及び BaCl ₂ シンチレータに関する研究)
WATANABE, Kohei (Saitama University)	Measurements of low-energy reaction cross sections for d + ¹² C at the Tandem accelerator facility (低エネルギー反応断面積測定に向けてー筑波大学タンデム加速器での d+ ¹² C 実験)
MIKAWA, Misaki	Development of Radio Isotope Beams for Unstable Nuclei ¹⁰ Be and ¹⁴ C using an Accelerator Mass Spectrometry System (加速器質量分析装置を用いた不安定核 ¹⁰ Be、 ¹⁴ C ビームの開発)
MITSUI, Maoto	Measurement of charge-changing cross section of proton-rich Ca isotopes (陽子過剰 Ca 同位体の荷電変化断面積の測定)

Undergraduate theses

NOGUCHI, Tomoki	Simulation of Interatomic Distance in the Solid Transmission Process of Heteronuclear Diatomic Molecular Ions (異核二原子分子イオンの固体透過過程における原子間距離シミュレーション)
INOUE, Daichi	Vibration Measurements Using Time-Averaged Holographic Interferometry (時間平均ホログラフィー干渉法による振動計測)
SAITO, Yoshihito	Development of radioactive ⁷ Be beams at the University of Tsukuba Tandem Accelerator Complex (筑波大学タンデム加速器施設における不安定核 ⁷ Be ビームの開発)

8.

LIST OF PERSONNEL

Tandem Accelerator Complex

K. Sasa	Director, Associate Professor
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T. Moriguchi	Assistant Professor
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T. Manako	Mechanical Engineer
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T. Yoshida	Electrical Engineer
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¹ The “research members” include the authors and coauthors within 5 years back from this fiscal year, as well as the members of research projects running at UTTAC.

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