JAEA Takasaki Annual Report 2006

(Ed.) Shigeru TANAKA

Takasaki Advanced Radiation Research Institute

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JAEA Takasaki Annual Report 2006

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JAEA Takasaki annual report 2006 describes research and development activities performed from April 1, 2006 to March 31, 2007 with Takasaki Ion Accelerators for Advanced Radiation Application (TIARA, four ion accelerators), and electron/gamma-ray irradiation facilities (an electron accelerator and three $^{60}$Co gamma-ray irradiation facilities) at Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency (JAEA Takasaki). These activities are classified into four research fields: 1) space, nuclear and energy engineering, 2) environmental conservation and resource security, 3) biotechnology and medical application, and 4) advanced materials, analysis and novel technology. This annual report contains 186 reports consisting of 178 research papers and 8 status reports on operation/maintenance of the irradiation facilities described above, and a list of publications, related press-releases, TV programs, patents, and the type of research collaborations as Appendices.


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高崎量子応用研究所研究年報 2006

日本原子力研究開発機構 高崎量子応用研究所
（編） 田中 茂

（2007年12月27日受理）

高崎量子応用研究所研究年報 2006 は、同研究所にある TIARA 施設（イオン加速器 4 基）及び電子・ガンマ線照射施設（電子加速器 1 基、^{60}Co ガンマ線照射施設 3 棟）を利用して2006年4月1日から2007年3月31日までの間に行われた研究・技術開発成果をまとめたものである。この研究年報には、1）宇宙・原子力・エネルギー、2）環境・資源、3）バイオ技術・医学応用、4）先端材料・計測・基盤技術の4分野に分類した178編の論文及び8編の施設の運転・管理状況報告からなる合計186編を収録する。また、論文リスト、新聞発表、テレビ放映、出願特許、及び研究実施形態・利用施設の一覧表を付録として含む。
PREFACE

This report covers research and development activities in Takasaki Advanced Radiation Research Institute, JAEA, during the period from April 2006 to March 2007, conducted with TIARA (Takasaki Ion Accelerators for Advanced Radiation Application), electron beam and Co-60 gamma-ray irradiation facilities.

One hundred eighty seven papers are appeared in this annual report in the fields of
1) Space, Nuclear and Energy Engineering,
2) Environmental Conservation and Resource Security,
3) Biotechnology and Medical Application,
4) Advanced Materials, Analysis and Novel Technology,
5) Status of Irradiation Facilities.

In the field of Space, Nuclear and Energy Engineering, the radiation resistance of solar cells and integrated circuits newly developed for space application has been evaluated using high-energy electrons, protons and heavy ions. Experimental and theoretical investigations of transient currents induced in typical device structures such as SOI (silicon on insulator) by energetic ions have been performed for clarifying mechanisms behind single event phenomena in order to develop prediction methods of the reliability of semiconductor devices, which will be used in radiation environment. Radiation resistant devices have also been developed using wide band-gap semiconductors like SiC and GaN. Radiation degradation of organic materials, e.g., insulating materials in superconducting coils of ITER, coaxial cables for J-PARC, and grease for the equipments of nuclear-cycle reprocessing have been examined for aiming at their practical usage. Radiation effects involving irradiation assisted stress corrosion cracking (IASCC) in structural materials of fusion, fast breeder and light water reactors have been intensively investigated using TIARA for securing reactor safety. In addition, high quality polymer electrolyte membranes have been fabricated using ion beams, electron beams and gamma-rays for their application to fuel cells.

In the field of Environmental Conservation and Resource Security, metal ion adsorbents have been developed with radiation grafting technique for removing toxic elements such as Cd, Pb and B in industrial waste water. R&D of graft adsorbents based on plant-derived polymers like polylactic acid (PLA) and carboxymethylcellulose (CMC) has been started. Radiation grafting technique has also been applied to improve the properties of materials for industrial use; low power-loss millimeter-wave planar antennas and high quality rubber wipers for automobiles have been successfully developed. Biodegradable hydro-gels and polymers from CMC and PLA have been investigated using radiation cross-linking, leading to the success in fabricating novel elastic CMC gels and elastic PLA materials with high heat resistance. Concerning environmental purification using electron beams and gamma-rays, new process technologies have been developed for the decomposition and removal of trace amount of pollutants such as formaldehyde in sterilization gas, dioxin wastes and endocrine disrupting chemicals.

In the field of Biotechnology and Medical Application, the ion beam breeding technique has been applied for many varieties of plants and microorganisms such as flower, crop, vegetable, yeast, alga and fungus. Functional analyses of radiation-response proteins of the radioresistant bacterium and the elucidation of characteristics of ion beam-induced damage and mutation have been progressed. Molecular mechanisms in radiation-induced bystander
effects were investigated in normal and cancer cells using heavy-ion microbeams in the aspects of initiators/mediators and the role of gap junction. The effect of ionizing radiation on the food-NaCl associative learning behavior in *Caenorhabditis elegans* was also investigated. Compartment model was applied to the positron imaging of a tobacco leaf and succeeded in obtaining parametric imaging of two photosynthetic processes, photoassimilation and sucrose-export. A new radionuclide for medical application, Br-76, was produced by using AVF cyclotron and examined for the diagnosis of cancer by positron emission tomography, PET. Clarification of the dynamics of anticancer drug was studied by analyzing trace elements in tumor cells using the in-air micro-PIXE system.

In the field of Advanced Materials, Analysis and Novel Technology, development of hydrogen gas sensor using tungsten trioxide (WO$_3$) has been conducted; it was founded that ion irradiation to WO$_3$ films improved their gasochromic coloration by introducing oxygen vacancies. In order to measure the three-dimensional distribution of elements in biological samples, such as carbon, hydrogen, and oxygen using the in-air micro PIXE system, STIM (Scanning Transmission Ion Microscopy) / CT (computed tomography) method was developed and introduced to the system; three-dimensional reconstruction of an insect’s head was successfully obtained using all tomograms with the system. All-permanent-magnet type ECR ion source (LECR) has been developed since 2003, and beam current of O$^{7+}$ was increased from 0.6 to 1.8 $\mu$A by the modification of the LECR in 2006. Negative Ge cluster ions were generated by a Cs sputter ion source, and Ge clusters up to Ge$_4$ at the beam current of nano ampere order was extracted from the source. Three-dimensional micro structures were fabricated with the Proton Beam Writing (PBW) technique using protons at two different beam energies of 1.2-and 3.0-MeV impinging on a layer of negative resist material. In the development of monitor and dosimeter for radiation, measurement technique of two dimensional dose distribution was developed by using B3 film dosimeters and a PC-scanner.

All the accelerators in TIARA, namely the AVF cyclotron, the 3-MV tandem accelerator, the 3MV single-ended accelerator and the 400-kV ion implanter, have been operated steadily as well as an electron accelerator and irradiation systems in Co-60 gamma-ray irradiation facilities. Especially, the cyclotron has been operated in this fiscal year providing ion-beams to all users perfectly without any cancellation of machine time, although there occurred 324 machine troubles. The total operation time of the cyclotron from the first-beam provision reached 50,000 h in April 2007, as the result of such continuous back-up efforts.

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Hideki Namba,  
Director General  
Takasaki Advanced Radiation Research Institute  
Japan Atomic Energy Agency
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Effect of Base-layer Carrier Concentration on Radiation Resistance of AlInGaP Solar Cell

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Optimum epitaxial layer structure of an AlInGaP solar cell as a top subcell in a triple-junction (3J) space solar cell has been studied. In 2006, the base-layer carrier concentration in AlInGaP single-junction (1J) solar cells was varied in order to clarify the suitable carrier concentration for radiation resistance of the cell.

AlInGaP 1J solar cell epitaxial wafers were grown by metal-organic vapor phase epitaxy (MOVPE). The carrier concentration in base-layer was varied for 1×10^{16} cm^{-3}, 3×10^{16} cm^{-3} and 3×10^{17} cm^{-3}. Solar cells were fabricated from the wafers. However, anti-reflection coating was not formed on the surface of the cells to avoid the effect of difference in reflectance. The cell size is 1cm×1cm. The cells were irradiated with 1MeV electrons (nitrogen ambient) and 3MeV protons (in vacuum) at room temperature.

Figures 1 and 2 depict the radiation degradation of the short-circuit current (Isc) and the open-circuit voltage (Voc) of the AlInGaP 1J solar cells due to 1MeV-electron and 3MeV-proton irradiations, respectively. In the case of degradation of Isc, there seems to be no significant dependency on the carrier concentration, while the cell with the concentration of 3×10^{17} cm^{-3} degrades greater than those with the concentration of the order of 10^{16} cm^{-3}.

These results indicate that the minority-carrier diffusion length in the p-AlInGaP base layer and its radiation degradation are not affected by the base-layer carrier concentration between 1×10^{16} cm^{-3} and 3×10^{17} cm^{-3}. On the other hand, the carrier removal effect due to the irradiation, which results in the decrease in built-in potential at the AlInGaP n/p junctions, is greater when the base-layer carrier concentration becomes in the order of 10^{17} cm^{-3}.

Reference
Semiconductor devices with Silicon On Insulator (SOI) technology are utilized for high-performance and low-power electronics such as computer servers for high-end commercial markets. SOI is a layered structure consisting of a thin layer of silicon mounted on an insulating substrate. The typical cross section image is shown in Fig.1. SOI technology is also very attractive as semiconductor devices for space application. SOI devices are expected to have an advantage over equivalent bulk technology devices for Single-Event Effects (SEEs) caused by heavy ions and high-energy protons because the sensitive volume for SEEs is much smaller than that of bulk Si devices. In JAXA, now we continuously perform the design and evaluation of radiation hardened logic circuits utilizing 0.15μm fully depleted SOI process technology \(^1\). This study is indispensable in order to develop next generation semiconductor devices for space application such as high-speed Microprocessors (MPUs) and Field Programmable Gate Arrays (FPGAs).

This year, several kinds of radiation hardened logic circuits such as latch and flip flops were designed and evaluated. In this report we describe the evaluation results of latch circuit as a representative case. Two types of radiation hardened latch circuit (Basic type and Optimized type) were designed and installed in the test chips. In general, Single Event Transient (SET) pulse is generated when an ion particle hit on the p-n junction of transistors. The propagation of this pulse noise leads to the malfunction of logic circuit (i.e. SEE). For SOI technologies, it is possible to eliminate the SET pulse generation on the output terminal by using circuit techniques \(^1\). Figure 2 shows the concept of the radiation hardened inverter logic circuits with redundant transistors. By using this, various types of radiation hardened logic circuit can be constructed. For the basic type latch circuit, SET-free inverter circuit is fully applied on the data input logic and the inverter loop. However the area, power consumption, speed penalties are too large compared to the conventional latch circuits. For the optimized type, some redundant p-type transistors are removed in order to minimize the penalty mentioned above. Test chips were fabricated with a 0.15 μm fully depleted SOI commercial process at OKI (Tokyo, Japan). Figure 3 shows the pictures of test device. In this chip, each kind of latch circuit composes the 4kbit memory array.

Kr and Xe ion irradiation test results are shown in Table 1. Some amount of SEU was observed only when Xe ion irradiated. It was confirmed that both types of latch circuits have a very good radiation tolerance. We plan to continue this study in order to construct the radiation hardened logic cell library for space SOI.

**Reference**


<table>
<thead>
<tr>
<th>Ion</th>
<th>Fluence [particles/cm²]</th>
<th># of Errors</th>
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<tr>
<td>Basic type</td>
<td>Optimized type</td>
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<tr>
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</tr>
<tr>
<td>Xe</td>
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</table>

*These errors may be attributable to considerable degradation caused by local total ionizing dose effect.
One of the most detrimental effects on semiconductor devices in radiation environments is single-event effects (SEEs). When a high-energy heavy ion strikes the device, electron-hole (e-h) pairs are generated along the ion-track and they lead to transient currents at the terminal of devices.

Musseau et al. proposed that the silicon-on-insulator (SOI) is one of the most promising structures to achieve higher SEE tolerant devices, because the charge collection can be suppressed by the existence of a buried oxide layer [1]. However, the anomalous charge collection in the SOI device was reported recently [2]. This fact suggests that dense charges generated under a buried oxide contribute to the charge collection. Therefore it is important to make clear the charge collection mechanisms through an insulation (oxide) layer due to heavy ion irradiation. In a previous study, we have measured transient currents induced through an oxide layer by ion incidence using metal oxide semiconductor (MOS) capacitors to understand the charge collection mechanisms in SOI devices. As a result, the transient current induced in MOS capacitors can be qualitatively interpreted in terms of displacement current [3]. However, the quantitative analysis has not yet done.

In this report, we perform numerical simulations on transient currents induced in MOS capacitors using an optimized physical model, and compare calculated results to experimental results.

The MOS capacitors used in this study are fabricated on p-type Si substrates. The concentration of boron acceptors in the substrate is $4.63 \times 10^{15} \text{ cm}^{-3}$. The oxide layer at a thickness of 31 nm was formed using dry oxidation at 1050 °C for 30 min. Aluminum was applied as the gate metal. Heavy ion irradiation experiments were carried out using the 3MV tandem accelerator at TIARA. The transient currents induced in the MOS capacitors by 15 MeV-oxygen ion incidence were collected by transient ion beam induced current (TIBIC) measurement system with single ion hit (SIH) system.

The transient currents were simulated by using the Technology Computer Aided Design (TCAD) version 10 provided by Synopsys. The DESS package in TCAD was used to simultaneously solve the Poisson and e-h continuity equations under the cylindrical symmetry (quasi-three dimensional) conditions. The following models were assumed in all simulations: (1) Fermi–Dirac statistics, (2) bandgap narrowing, (3) mobility model dependent on doping concentration, and carrier density, (4) high field saturation, and (5) incomplete ionization. In addition, since the ion initial track structure strongly affects the transient current, the distributions of e-h pairs are calculated by considering the $\delta$-electron production with MeV ion penetration and keV $\delta$-electron transmission theory, instead of the cylindrical track structure generally accepted in TCAD simulations.

Figure 1 shows the comparison of the experimental results with the simulated results. The total collected charges do not show strong bias dependence and slightly decrease with increasing bias. On the other hand, as for the peak currents, the values increase with increasing bias. As a result, the peak currents and the total collected charges obtained by time integration of transient currents agree well with the measured values, errors being less than 10%.

Fig. 1 Comparison of the experimental results with simulated results. Bias dependence Peak Current and Total Collected Charge.

References
Heavy-ion Induced Current in Silicon-on-insulator Based Devices

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In order to clarify the charge collection mechanism through a buried oxide layer in SOI structure, the single heavy-ion induced transient gate current in MOSFET has been investigated. The transient gate current with both positive and negative peaks is observed by hitting an ion to gate area, and the amount of collected charge converges on zero within 100ns after irradiation. The transient current in n-MOSFET is larger and also shorter than p-MOSFET because of mobility difference between electrons and holes. It is also confirmed that the current peak increases with applied drain voltage during irradiation.

宇宙環境下で半導体デバイスを使用する場合、重イオンなど高速荷電粒子の照射により誘起される電流に起因したシングルイベント現象が問題となる。能動領域の薄膜化が可能なSOI (Silicon on Insulator) デバイスは、埋め込み酸化膜の存在により能動領域が限定されているため、重イオン照射誘起電流の評価に寄与する。本研究では、酸化膜を介した電荷収集機構について検討を進めてきた2)。今回はpチャネルおよびnチャネルMOSFETに対する照射実験を行い、電流発生機構について検討を行った。

Alゲートp-MOSFETおよびn-MOSFET（ゲート酸化膜厚d_ox=40 nm, ゲート長L=100 µm, ゲート幅W=300µm）を作製し、日本原子力研究開発機構の重イオンマイクロビームシステムを使用して酸素イオン（15MeV）の照射を行った。なお、照射による損傷を軽減し、かつ単一イオンの入射に起因した誘起電流を正確に測定するため、Single Ion Hitシステムを用いて単一イオン照射を実施し、高速オシロスコープにより照射誘起過渡電流を測定した。

図1に、ソースドレイン電極を接地した状態で、MOSFETゲート領域に重イオンを照射した際の、ゲート電極における照射誘起電流および収集電荷量（電流積分値）を示す。なお照射中のゲート印加電圧は、p-, n-MOSFETそれぞれ-10および+10 Vであり、ゲートから電源方向へ流れ出す電流を正方向として表示した。結果より、照射後1〜2 ns程度で酸化膜印加電界分である銭電流ピークが、その後、反方向の小さな電流がそれぞれ観測されること、また、収集電荷量は照射後数10nsで0に収束することがわかった。以上の結果より、酸化膜を介した重イオン照射誘起電流は、変位電流によるものであることを確認した。p-MOSFETの場合、照射により基板内で発生した正孔の酸化膜表面での蓄積による酸化膜電界の急速な変化により大きな正方向電流が流れ、これら過剰正孔のドレインソースによる収集により初期状態に戻る際に、逆方向電流が発生したものと考えられる。また、n-MOSFETの方が短時間・大振幅の電流ピークとなった。これは、正孔に比べて電子の移動度が高いことに起因する。

また、照射誘起ゲート電流のドレイン電圧依存性の評価を行った。その結果、pチャネル、nチャネル形共に、電流ピーク値は照射中に印加したドレイン電圧（絶対値）と共に、大きくなることがわかった。ドレイン電圧印加時には、ドレイン近傍のゲート酸化膜電界はソース近傍に比べて小さくなる（表面電位が大きくなる）。ただし、重イオンが入射されると、照射により発生した高密度の正孔が内部電界により基板表面に引き寄せられ、帯電部分全体に蓄積されるため、表面電位は場所によらずほぼ同電位になることが予想される。すなわち、ドレイン近傍の酸化膜電界の変位速度は、ドレイン電圧と共に大きくなるものと考えられる。以上のメカニズムにより、重イオン照射誘起ゲート電流のドレイン電圧依存性を説明できることがわかった。

Fig.1 Heavy-ion induced gate currents (solid lines) and collected charges (dashed line) in p- and n-MOSFET.

References
1-05 Bias Dependence of the Single Event Transient Currents due to Nuclear Products in Semiconductors II

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High energy neutrons and protons can trigger Bit Error Rate (BER) degradation in optical data links used in radiation environments. 1,2) It is known that the photodetectors, which are the principle component of optical receivers, are more prone to BER degradation than external encoding electronics. Although the direct ionization results in BER degradation in the photodetectors operated under low optical power level, it is also possible for the secondaries generated by nuclear reactions to induce BER degradation. In silicon based materials, the inelastic as well as elastic reactions generate high-energy secondaries such as He, C, N, O, F, Ne, Na, Mg, Al, Si, P, and recoiling Si atoms with energies up to several tens of MeV. The ionizing energy loss of these secondaries induces a current on the terminal of the photodetector resulting in a Single Event Transient (SET) current that contributes to BER degradation.

In order to clarify the carrier dynamics resulting in the SET currents in Si pin photodiodes due to neutrons, we have measured the SET current induced by energetic heavy ions (C, O, and Si ions) assuming secondaries. Then we have evaluated the bias dependence of the SET characteristics such as collected charge, peak current, fall time and rise time, respectively.

The device examined in this work was a commercial 1.5 GHz (at -3 dB) Si pin photodiode with a diameter of 450 μm. The p-layer width was about 0.2 μm, the depletion width was about 15 μm at -10 V as estimated by the capacitance-Voltage (C-V) and cross-sectional Transient Ion Beam Induced Current (TIBIC) methods. All proton induced SET currents were measured at various biases from -5 to -30 V.

Figure 1 (a, b, c) show the bias dependence of the collected charge, the peak current, the fall time, and the rise time due to C, O, and Si ions with energy up to 15 MeV, respectively. As shown, the charge increases in proportion to ion energy for all ions and does not depend on ion energy. The peak current increases with increasing applied bias although fall time and rise time decrease. These facts show that the higher bias leads to the faster drift velocity of carriers, which results in the higher peak current and shorter fall and rise time. On the other hand, the peak current decreases with increasing atomic number. The most probable reason is that the charge screening effect is influenced considerably when the atomic number increases. 3)

References

1) E. Petersen, IEEE NSREC Short Course, section III (1997)
2) S. Onoda et al., JAEA Takasaki annual report 2006 (2007) 9

Fig.1 Bias dependence of the SET current characteristics such as the collected charge, the peak current, the fall time, and the rise time induced by (a) C, (b) O, and (c) Si ions with energy up to 15 MeV.
Study of Charge Induced in 6H-SiC n\textsuperscript{+}p Diodes by Gold Ion Incidence

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Silicon carbide (SiC) is expected to be applied to particle detectors used in radiation environments since SiC has superior radiation tolerance\textsuperscript{1).} For this purpose, it is important to clarify the behavior of charge generated in SiC by ion incidence. In a previous study\textsuperscript{2)}, it was reported that the Charge Collection Efficiency (CCE) obtained from 4H-SiC diodes irradiated with alpha particles was 100 %. The CCE of 100 % was also reported for 6H-SiC diodes irradiated with oxygen (O) ions\textsuperscript{3).} These results suggest that SiC is suitable for particle detectors. For the development of particle detectors, the CCE of SiC diodes should be evaluated using a wide variety of ions. In this study, we evaluate charge generated in 6H-SiC diodes by 12 MeV-gold (Au) ions.

The samples used in this study were 6H-SiC n\textsuperscript{+}p diodes fabricated on 30 μm thick p-type epitaxial layer with an aluminum (Al) concentration of 8x10\textsuperscript{14} - 1x10\textsuperscript{15}/cm\textsuperscript{3}. The epitaxial layer was grown on a p-type substrate (3.5° off, Si-face) by chemical vapor deposition. The 100nm thick n\textsuperscript{+} region was formed using phosphorus (P) ion implantation (a mean P concentration of 5x10\textsuperscript{19}/cm\textsuperscript{3}) at 800 °C followed by annealing at 1650 °C for 5 min in argon (Ar). Al electrodes were formed using a liftoff technique. The diodes were irradiated with 12 MeV-Au microbeams under applied reverse biases, and transient current generated by irradiation of Au ions was measured using the Transient Ion Beam Induced Current (TIBIC) system\textsuperscript{4).} Figure 1 shows the value of collected charge as a function of applied reverse bias. The estimated value of charge generated in the depletion layer (ideal value) is also represented as a broken line. In the estimation, ionizing energies deposited from 12 MeV Au ions into SiC were calculated by Monte Carlo simulation code SRIM2003\textsuperscript{5).} The energy for the generation of an e-h pair (E\textsubscript{e-h}) in 6H-SiC is assumed to be 7.8 eV (= 2.8E\textsubscript{e-h}) on the analogy of E\textsubscript{e-h} in Si since the value of the energy for 6H-SiC has not yet been determined. The collected charge of 0.1 pC is obtained and the value does not depend on the applied biases. On the other hand, the ideal value is estimated to be 0.195 pC. Since the depletion layer is estimated to be 5 μm at an applied bias of 20V and the range of 12 MeV-Au is 1.9μm, the obtained result indicates that approximately half of charge generated in the depletion layer is not collected despite under the reverse biases.

The CCEs for 6H-SiC diodes irradiated with 12 MeV-O, silicon (Si), nickel (Ni) ions were also investigated. In the case of O and Si ions, the CCE obtained for this diode showed more than 95 %. On the other hand, the values of the CCE for Ni- and Au-irradiated diodes are 86 and 53 %, respectively. The distributions of e-h pairs were calculated using the method developed by Kobetch and Katz (KK)\textsuperscript{6,7).} As the result, the estimated result indicates that the density of e-h pairs in Au-irradiated SiC is 100 times higher than that in O-irradiated SiC. Therefore, the decrease in the CCE by Au irradiation obtained in this study can be interpreted in terms of the recombination of carriers generated in depletion layer because the recombination of generated carriers might occur in such high dense plasma.

This study was partially supported by the Ministry and Education, Science, Sports and Culture, Grant-in-Aid for Scientific Research (B), 2006, 18360458.

References
EPR Identification of Intrinsic Defects in 4H-SiC: Positively-charged Carbon Antisite-vacancy Pairs

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To improve the radiation hardness of SiC devices, identification of radiation-induced defects are required. We use electron paramagnetic resonance (EPR) technique for microscopic structural identification of intrinsic defects in 4H-SiC. To track the angular dependence of the $^{29}\text{Si}^{13}\text{C}$ hyperfine (HF) lines which does lead to decisive structure determination, it is desirable to produce the intrinsic defect of interest selectively at sufficient concentration. We took a sort of defect engineering approach by using various combinations of the temperature and the dose of 2 MeV electron irradiation ($1 \times 10^{17}$–$1 \times 10^{19} \text{e/cm}^2$), the temperature (400–1200 °C) of heat-treatment after irradiation, and 4H-SiC samples ($3 \times 10 \times 1.5 \text{mm}$) of both $n$- and $p$-type of different dopant concentrations.

Carbon antisite-vacancy pair $C_{\text{Si}}V_{\text{C}}$, which is one of nearest neighbor carbon atoms moving into the vacant silicon site with a carbon vacancy left, corresponds to a bistable counterpart of silicon vacancy (Fig.1). The negative silicon site with a carbon vacancy left, corresponds to a nearest neighbor carbon atoms moving into the vacant silicon site. Thus, $C_{\text{Si}}V_{\text{C}}$ is a dominant carrier-compensating, intrinsic defect producing the SI properties. Theoretical studies predicted that the positive charge state $[C_{\text{Si}}V_{\text{C}}]^+$ should be much more stable than silicon vacancy in $p$-type while $[C_{\text{Si}}V_{\text{C}}]^-$ should be metastable in $n$-type.$^{2,3}$

In the present work, the positive charge state $[C_{\text{Si}}V_{\text{C}}]^+$ ($S=1/2$) has been identified in electron-irradiated $p$-type (B-doped) 4H-SiC ($2 \times 10^{18} \text{e/cm}^2$, 800 °C, RT carrier concentration before irradiation $\sim 10^{15} \text{cm}^{-3}$).$^6$ The EPR spectra labeled $\text{HEI9a/b}$ and $\text{HEI10a/b}$ (Fig.2) were recorded at 295K on a Bruker E500 X-band spectrometer.

In 4H-SiC, both silicon and carbon atoms have two inequivalent lattice sites, hexagonal ($h$) and quasi-cubic ($k$) sites. For each of four spectra, $\text{HEI9a/b}$ ($C_{\text{Si}}$) and $\text{HEI10a/b}$ ($C_{\text{Si}}$, the $^{13}\text{C}$ ($I=1/2$, natural abundance 1.1 %) HF tensor has been determined from the fitting of the angular dependence of the HF lines (See Fig.2 for $b/c$). The $^{13}\text{C}$ HF tensors obtained are useful to assign each of four spectra to one of ($hh$, $kk$, $hk$, $kh$) configurations by comparing with the HF tensors of four configurations estimated by ab initio supercell calculations.

Observation of four sets of spectra, which all exhibit similar dose dependence and thermal behavior (Fig.3) and are assignable to four configurations ($hh$, $kk$, $hk$, $kh$), respectively, is a strong basis that $HEI9/10$ is arising from $[C_{\text{Si}}V_{\text{C}}]^+$. We apply photo-EPR technique which measures the EPR signal intensity as a function of the wavelength of illuminating light, to determine the defect level in the band gap. It has been determined that the electronic level of $C_{\text{Si}}V_{\text{C}}^+$ is located at around $E_{V^+}+1.4\text{eV}$.

References
Mechanisms of Decrease in Hole Concentration in Al-doped 4H-SiC by Irradiation of 200 keV Electrons

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From the temperature dependence of the hole concentration $p(T)$ in Al-doped 4H-SiC epilayers irradiated with 4.6 MeV electrons, we reported that the density $N_{\text{Al}}$ of a shallow acceptor with $E_v + 0.2$ eV, which is an Al atom at a Si sublattice site, was significantly reduced, while the density $N_{\text{Defect}}$ of a deep acceptor with $E_v + 0.35$ eV was slightly decreased, as shown by triangles of Fig. 1. Here, $E_v$ is the valence band maximum. In unirradiated epilayers, $N_{\text{Defect}} = 0.6 N_{\text{Al}}$ in a range of $N_{\text{Al}}$ between $8 \times 10^{14}$ and $5 \times 10^{16}$ cm$^{-3}$, as shown by open symbols of Fig. 1.

Since electrons with $<0.3$ MeV can displace only carbon (C) atoms in SiC whereas electrons with $>0.5$ MeV displace all the atoms (i.e., C, Al and Si) in SiC, we investigate the changes of $N_{\text{Al}}$ and $N_{\text{Defect}}$ in a 10 μm-thick Al-doped 4H-SiC epilayer by irradiation of 200 keV electrons. After the Hall-effect measurement was carried out in the epilayer irradiated with $1 \times 10^{16}$ cm$^{-2}$ fluence, the epilayer was irradiated with $2 \times 10^{16}$ cm$^{-2}$ fluence. Figure 2 shows $p(T)$ denoted by open circles (unirradiated), solid circles (fluence: $1 \times 10^{16}$ cm$^{-2}$) and solid diamonds (total fluence: $3 \times 10^{16}$ cm$^{-2}$). At low temperatures, $p(T)$ decreases with increasing fluence, whereas $p(T)$ seems unchanged at high temperatures, indicating that by irradiation of 200 keV electrons $N_{\text{Al}}$ is decreased while the sum of $N_{\text{Al}}$ and $N_{\text{Defect}}$ is unchanged.

From the analysis of $p(T)$, the values of $N_{\text{Al}}$ and $N_{\text{Defect}}$ were determined, and are shown as an open circle (unirradiated), a solid circle (fluence: $1 \times 10^{16}$ cm$^{-2}$) and a solid diamond (total fluence: $3 \times 10^{16}$ cm$^{-2}$) of Fig. 1. Different from the changes of $N_{\text{Al}}$ and $N_{\text{Defect}}$ by irradiation of 4.6 MeV electrons, $N_{\text{Al}}$ decreases with increasing fluence of 200 keV electrons, while $N_{\text{Defect}}$ increases. Moreover, the decrement of $N_{\text{Al}}$ is nearly equal to the increment of $N_{\text{Defect}}$. Therefore, the displacement of only C atoms by irradiation of 200 keV electrons is considered to change the Al acceptor into the deep acceptor.

The changes of $N_{\text{Al}}$ and $N_{\text{Defect}}$ by irradiation at more fluences ($5 \times 10^{16}$ and $7 \times 10^{16}$ cm$^{-2}$) of 200 keV electrons are now investigated.

References

Fig.1 Relationship between $N_{\text{Al}}$ and $N_{\text{Defect}}$. The open symbols of squares, circle and triangle are the relationship before irradiation. The solid circle and diamond represent the relationships after irradiation of $1 \times 10^{16}$ and $3 \times 10^{16}$ cm$^{-2}$ with 200 keV electrons, respectively. The solid triangle represents the relationship after irradiation of $2.6 \times 10^{14}$ cm$^{-2}$ with 4.6 MeV electrons.

Fig.2 Temperature dependence of hole concentration. The open circles represent $p(T)$ before irradiation, and the solid circles and diamonds represent $p(T)$ after irradiation of $1 \times 10^{16}$ and $3 \times 10^{16}$ cm$^{-2}$ with 200 keV electrons, respectively.
1-09 First-principles Molecular Dynamics Simulation of SiO$_2$/SiC Interface of Silicon Carbide Devices

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Silicon carbide (SiC) semiconductor devices are expected to be used in severe environments such as outer space and nuclear power plants. The performance of the SiC MOSFET device is below theoretically expected levels because the defect in SiO$_2$/SiC interface degrades the electrical performance of the device. The relation among atomic structures, interface defects and electrical characteristics is not clear at present. In this study, we tried to solve these problems by the computer simulation of electronic structure of the SiO$_2$/SiC interface. It is important to construct an amorphous SiO$_2$ ($\alpha$-SiO$_2$) structure on SiC to emulate the real device interface structure by the computer simulation because the oxide layer of the real device is an amorphous structure. We aimed to generate $\alpha$-SiO$_2$/SiC structure with large-scale (1000 atoms scale model) calculation to which the amorphous structure was able to be constructed.

Amorphous SiO$_2$/SiC interface structure is made by applying the heating and quenching calculation that uses the first-principles molecular dynamics code VASP$^{1)}$ (Vienna ab-initio Simulation Program) to the atomic interface structure model that consists of SiC crystal substrate and crystalline SiO$_2$ thin layers. The heating and quenching calculation of 444 atoms of SiO$_2$/SiC interface structure that had been done last year used the condition of 3.0 ps heating by 4000 K and the cooling speed of -1000 K/ps. It was confirmed that the SiO$_2$ layers of the interface structure were almost amorphous structure.$^{2,3)}$ The atomic density of the interface structure was obtained in the direction of the perpendicular of the interface. (Fig.1) In the SiO$_2$ layers near the interface, a periodic change of the density that reflects the crystalline structure was not seen; therefore, these layers are good amorphous structures. However, the density of the SiO$_2$ layers near the surface shows a periodic density change. It is guessed that the recrystallization occurred from the surface in the quenching process because the crystalline structure remained near the SiO$_2$ surface. It was suggested that the continuance annealing is necessary.

The generation of $\alpha$-SiO$_2$/SiC interface structure was tried by using the large scale model that consisted of 1017 atoms.

References
Development of Optoelectronic Devices for Radiation Environments - Optical Gain of Eu Implanted AlGaN -

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Temperature dependent luminescence and the optical gain of AlGaN doped with Eu ions are investigated for demonstrating the capability of rare-earth doped light emitting devices for radiation and/or hard environments. The Eu related photoemission, caused by 4f inner shell transitions, shows excellent thermal stability and even at 400K the luminescence intensity is more than 50% for low temperature (10K) case. Optical gain and loss measurements are carried out using variable stripe length and shifting excitation spot methods, respectively. The estimated optical gain is as high as 100 cm⁻¹, which is comparable to that reported molecular-beam-epitaxy grown GaN:Eu sample.

Fig.1 Optical gain measurement for Al0.3Ga0.7N:Eu at 300K. The Eu dose was 1×10¹⁵ cm⁻². The excitation power density was ~1 MW/cm².

References
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Gamma Radiation Effects on GaN Based Electron Devices

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GaN electronic devices, especially AlGaN/GaN Heterostructure Field Effect Transistor (HFETs), are expected for high-frequency and high-power applications due to wide-bandgap and III-V semiconductor characteristics 1, 2. Since the application includes power module and microwave power transmission for space solar power system, the employment of GaN-based devices in artificial satellite in space is being considered. The purpose of this research is to investigate the tolerance of these devices under γ-ray irradiation in space and other harsh environment 3-5.

The sample AlGaN/GaN HFETs were fabricated on (0001) sapphire substrate. It consisted of a buffer layer, a 3 µm undoped GaN layer, a 6 nm undoped AlGaN barrier, a 12 nm Si-doped AlGaN layer with doping concentration of \(4 \times 10^{18} \text{cm}^{-3}\), and a 6 nm undoped AlGaN barrier layer (Fig. 1). The Al mole fraction of all the AlGaN layers was 25%.

Ohmic contact was formed using lift-off technique with electron-beam-evaporated Ti/Al/Ni/Au (50/200/40/30 nm). They were annealed at 850 ºC for 1 minute in N₂.

The samples were irradiated with Co sources at JAEA Takasaki. The total dose of \(γ\)-ray was 8.9 Mrad and 86.7 Mrad at the dose rate of 1x10⁶ rad/h. The channel length and width of the measured transistor was 4 µm and 50 µm, respectively.

If the FETs are damaged, variations will appear in the gate leakage current, threshold voltage or carrier drift mobility. As shown in Fig. 2, each sample has inherently different gate leakage currents, but they did not change after the irradiation. Also, the threshold voltages did not change. Only the difference was the increase of drain current (\(I_D\)).

Converting drain voltage (\(V_D\)) versus drain current characteristics into drain voltage versus drain conductance (\(G_D\)) characteristics, it was found that the variation was only in low drain voltages and a constant resistance variation regardless of the gate bias. From these facts, the variation is attributable to improvement in the ohmic contact by \(γ\)-ray irradiation. The annealing in the fabrication process may have been insufficient.

Radiation experiments under a DC bias condition were done on different Metal-Insulator-Semiconductor (MIS) HFET. The FET has 10nm SiN on AlGaN layer deposited by sputtering. DC bias of -9V was applied to the gate electrode while the drain terminal was kept open.

After irradiation of \(γ\)-ray at 1 Mrad, no particular variation was observed except for small ohmic contact improvement similar to the previous experiment (Fig. 3).

Although material and process technologies for AlGaN/GaN HFETs have not yet been well controlled and the performance variations exist even without radiation, no particular degradation was observed even under extremely high dose irradiation of 87 Mrad.

References

Fig. 1 Schematic cross section of sample AlGaN/GaN HFET.

Fig. 2 Gate leakage current before and after irradiation of \(γ\)-rays at 86.7 Mrad and 8.9 Mrad. No bias was applied. Drain to source voltage(\(V_{ds}\)) was zero.

Fig. 3 \(I_D-V_D\) and \(G_D-V_D\) characteristics of the MISHFET, before and after 1 Mrad irradiation under the gate bias of -9V.
In this report, the impact of radiation damage on the device performance of 4H-SiC Metal Semiconductor Field Effect Transistors (MESFETs) and GaAlAs Light Emitting Diodes (LEDs) are investigated under different fluence and fluence rate conditions.

4H-SiC MESFETs were fabricated on 4H-SiC semi-insulating wafers with an epitaxial layer produced by CREE. The length and width of the gate were 0.5 and 100 μm, respectively. GaAlAs-double hetero junction-LEDs (Sharp GL3UR44) with light wavelength 660 nm were also used in this study.

Electron irradiations into the samples were done at room temperature using the 2-MV electron accelerator at JAEA, Takasaki. The used electron fluence ranged from 1 x 10^{13} to 1 x 10^{17} e/cm². Before and after irradiation, the input drain-source current/Gate-source voltage characteristics (I_DS-V_GS) of the SiC-MESFETs and I/V characteristics of the GaAlAs LEDs were measured with a parameter analyzer (HP-4156).

To investigate the recovery behavior due to annealing of radiation damage, isochronal thermal annealing was carried out for 15 min at temperatures between 100 and 300 °C under nitrogen flow without bias.

Figure 1 shows the annealing rate of I_DS as a function of annealing temperature for SiC-MESFETs irradiated with electrons at 5 x 10^{16} e/cm². Assuming that the recovery process is a first order reaction, the activation energy of the recovery is estimated from Fig.1 to be 0.27 eV.

Figure 2 shows the typical results of I/V characteristics for different electron fluence rates (4.68 x 10^{11} and 4.68 x 10^{12} e/cm²/sec). Total fluence was selected for 1 x 10^{15} e/cm². From this figure, it is noted that the remarkable degradation is observed for low dose rate condition than for high rate irradiation. This fluence rate dependence of degradation can be explained in terms of the heat impact in bulk crystal during irradiation.

References
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A laminated material composed of glass cloth / polyimide film / epoxy resin will be used as an electrical insulating material in the superconducting coil of the International Thermonuclear Experimental Reactor (ITER). This material is fabricated by impregnation of epoxy resin into a glass cloth / polyimide film prepreg and subsequent curing. Therefore, the viscosity and pot-life (time to cure the resin) of the epoxy raw material are very important for the fabrication of the insulating material. In addition, sufficient mechanical and electrical properties are required. Moreover, the insulating material must show high radiation resistance and thermal resistance, because it will be exposed to severe environments such as high radiation field and low temperature of 4K.

In this work, the gas evolution from laminated materials by gamma ray irradiation at 77K was investigated, and the difference of gas evolution behavior due to difference of components in the epoxy resin was discussed.

Two kinds of the laminated materials with different components of epoxy resin were investigated (Table 1). Each sample was put into a glass tube, and the glass tube was evacuated and sealed. The samples were irradiated by gamma rays up to dose of 30 MGy as maximum at a dose rate of 37.5 kGy/h at 77K, and then the gas analysis was conducted by a gas chromatograph at room temperature after storage of the samples at room temperature for 16 hours.

Figure 1 shows the evolved gases from the laminated materials by the irradiation of gamma rays at 77K. The total amount of evolved gas increased almost linearly with increasing in the dose. Main components of the evolved gases were hydrogen (H₂), carbon monoxide (CO) and carbon dioxide (CO₂). Hydrocarbon gases such as methane (CH₄) were evolved as minor components with an order of 10⁻⁸⁻¹⁰⁻⁷ mol/g. We have investigated the gas evolution from a polyimide film by the irradiation of gamma rays at 77K¹. It was found that the total amount of evolved gases was in an order of 10⁻⁹ mol/g. Therefore, the gas evolution from the laminated materials shown in the Fig. 1 is attributed by decomposition of the epoxy resin. Comparing the sample A with the sample B, the amount of the evolved gases from the sample A containing cyanate ester was about 60 % less than that from sample B containing TGDDM. This suggests that the insulating material for the superconducting coil with low gas evolution can be obtained by using the cyanate ester as a raw material for epoxy resin.

Reference
Gamma-ray Irradiation Test of Instrumentation and Operational Amplifier for Remote Maintenance System of ITER

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Components in the vacuum vessel of the ITER, an experimental fusion reactor to be constructed under international cooperation from this year, must be replaced remotely for maintenance because neutrons created by D-T fusion reactions activate structural materials. The radiation environment during the maintenance operation is about 0.5 kGy/h and therefore, the total dose is 1 MGy if the maintenance operation takes three months. The irradiation test has been performed for the components of the ITER remote maintenance system\textsuperscript{1,2)} and the present report describes the irradiation test of the instrumentation and operational amplifiers for strain gauges in order to examine which amplifier on the market can be used from the viewpoint of radiation-hardness. The tested amplifiers include the rad-hard products whose guaranteed acceptable dose is about 1 kGy.

Table 1 shows the list of tested instrumentation and operational amplifiers. Bipolar amplifiers were mainly selected because they are expected as rad-hard from the theoretical point of view. The dose rate in the irradiation test was almost the same value as that in the real environment of the ITER maintenance operation, 0.5 kGy/h. The amplifiers were irradiated for a certain period with bias current and after that they were took out from the cell for the measurement of their electrical characteristics such as input bias current, offset voltage, gain and so on. This cycle of irradiation and measurement was repeated.

First, a preliminary irradiation test was performed with the total dose of 20 kGy for screening. As a result, three kinds of amplifiers (AD524CD, HA1-5104-2 and HS1-5104ARH-Q) still operated normally although some of characteristics were slightly out of specification. Regarding other amplifiers, AD623BR and LT1078CN8 malfunctioned at 0.25 kGy. One of rad-hard amplifiers, RH1078MH, had functioned until 2 kGy, which is higher than the guaranteed value of 1 kGy, with some of characteristics deviating from specification and malfunctioned at 9 kGy.

Next, the irradiation test was performed until 417 kGy for the three amplifiers which functioned in the preliminary test. The instrumentation amplifier, AD524CD, had functioned until 56 kGy with some of characteristics deviating from specification, and malfunctioned at 169 kGy. The normal operational amplifier, HA-5104-2, had functioned until 169 kGy with some of characteristics deviating from specification, and malfunctioned at 417 kGy. The rad-hard operational amplifier, HS1-5104ARH-Q, had functioned without deviation of characteristics from specification until the guaranteed value, 1kGy, and had continued to function from 2 kGy till 417 kGy with some of characteristics deviating from specification. Figure 1 shows the transition of the open loop gain of HS1-5104ARH-Q during irradiation. The broken and dotted lines show the reference sample and specification, respectively. The gain showed the minimum value around 10 kGy, recovered after that and decreased again from 169 kGy. The deviation from specification is generally limited and therefore, this amplifier can be used until 0.4 MGy exceeding the guaranteed value, 1 kGy. The normal operational amplifier can be also used if it is replaced enough frequently. Regarding HS1-5104ARH-Q, its acceptable total dose will be investigated in the future study.

References

<table>
<thead>
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<th>Name</th>
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<tr>
<td>AD623BR</td>
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<tr>
<td>AD524CD</td>
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<td>Op.</td>
<td>1 kGy</td>
</tr>
<tr>
<td>LT1078CN8</td>
<td>Linear Technology</td>
<td>Op.</td>
<td>No</td>
</tr>
<tr>
<td>RH1078MH</td>
<td>Linear Technology</td>
<td>Op.</td>
<td>1 kGy</td>
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Fig. 1 Open loop gain of HS1-5104ARH-Q
Development of Radiation Resistant O-ring for Vacuum Use

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A superconducting magnet system is required for J-PARC Neutrino beam line to bent the 50 GeV 750 kW proton beam. The magnet system consists of about 150 m long vacuum vessel system to provide the thermal isolation for the superconducting magnet. The vacuum vessel system requires a lot of o-rings, of which size can be as large as 1 m diameter. Conventional o-rings, which uses the materials like fluoroelastomer (FKM), do not have sufficient resistance against radiation for long term use. High radiation resistant o-rings, such as metal o-rings, have demerits in costs or handling. Similar problems exist at the expansion joint sealants of the J-PARC accelerator tunnels. JAEA and Hayakawa Rubber Co. Ltd. jointly develop the new sealants based on the ethylene propylene diene monomer (EPDM) rubber. The new o-ring that can resist the irradiation up to 1 MGy has been developed jointly by KEK, JAEA, and Hayakawa Rubber Co. Ltd. based on the EPDM similar to that used for the expansion joint sealant. This report summarizes the irradiation tests performed for this o-ring material.

The samples were irradiated with γ-ray at the Cobalt-60 irradiation facility in Taka saki Advanced Radiation Research Institute, JAEA. The irradiations were made up to 5 MGy at room temperature with air environment. The mechanical tests based on the Japanese Industrial Standard (JIS) were performed for the samples. The mechanical tests include measurements of hardness (JIS-K6253), tensile stress (JIS-K6251), and compression set (JIS-K6262). The measurement was made for the samples irradiated to 0, 1, and 3 MGy. The out gas measurements of the samples were also made for the samples irradiated to 0 and 1.25 MGy.

The measurement results of the compression set are shown in Fig. 1. The FKM sample was destroyed after 1 MGy irradiation and no data was available. All the EPDM samples show good results not only in compression set but also all the mechanical measurements, indicating that the EPDM can be used as o-rings up to 1 MGy irradiation.

The results of the out gas measurement are shown in Fig.2. The results indicate that one of the EPDM sample (517) shows a good performance. We selected the sample 517 as the final candidate from the measurement results shown here as well as the good processability. The final products use the same EPDM as 517 but with small modification of final process. The EPDM was renamed to 517LT, and were tested for both mechanical and out gas measurements. The mechanical measurements results show better performance than those of 517. The out gas measurement results were not as good as the one for 517, but still acceptable from the requirement.

The o-rings are now in the stage of mass production and will be installed in the magnet system. The magnet system will be completed in the end of 2008 and operated in 2009.

Reference

Fig.1 JIS-K6262 Compression set measurement results. No.509 is the FKM sample, which destroyed at 1 MGy irradiation. The other samples are EPDM based.

Fig.2 Out gas measurement results, BaseVT and the P-509 are the FKM, the others are EPDM. The material chosen for the final products is 517LT.
Measurement of Electrical Property Change in Commercially Available Coaxial Cable by Gamma-ray Irradiation

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Introduction

Coaxial cable is an essential element for a communication tool in a precise measurement system because of stable characteristic impedance and electrical noise proof performance. The J-PARC 1), a research facility complex with high-intensity proton accelerators, apply a plenty amount of coaxial cables to make good telecommunications regarding to the proton beam circumstance, the accelerator status, RF signal control and the safety issues. The proton beam power of the J-PARC is up to 1 MW on the target, which introduces the high radiation area along the proton beam line and close to the target region caused by undesirable beam loss of the accelerators. We measured the electrical property change in the commercially available coaxial cable under the high dose rate gamma-ray irradiation to confirm the reliability of the control and the data acquisition tool of the J-PARC.

Measurement

A pair of 40 m long coaxial cables (Fujikura NH-8D-SFA) was prepared as a race-track shape to compare the difference between a fresh cable and an irradiated cable. The irradiated cable is 3 MGy pre-dosed by gamma-ray. Both cables are tied up and installed in the cell facing the Co-60 gamma-ray source on even dose condition, and the characteristics are measured alternatively. The dose rate of the gamma-ray is chosen 6.3 kGy/h to simulate the high radiation spot in the J-PARC. The measured items are insulation resistance, RF attenuation, phase shift, time delay, charge up and characteristic impedance.

Result

1) No gamma-ray run as static characteristics

Slightly degradation, range of a few percent, was observed for RF attenuation, phase shift, time delay and characteristic impedance on the 3 MGy pre-dose cable caused by increasing capacitance and dielectric constant shift at the insulator material. The insulation resistance between the inner conductor and the outer conductor decreased in the pre-dose cable by 20 % of the fresh one. The absolute value of the resistance still remains at the normal use level.

2) High gamma-dose run as dynamic characteristics

In the high radiation condition, the property of the pre-dose cable shows a constant deterioration for phase shift, time delay and characteristic impedance by several percent. The RF attenuation was -40 dB/km at GHz band. The charge up phenomenon was observed both the fresh cable and the pre-dose cable. Potential difference between the inner conductor and the outer conductor was measured during the gamma-ray irradiation period. The charge up voltage fluctuated through the beginning to the several minutes after the irradiation period 2). These phenomena affect like a common mode noise to the control and the beam monitoring system of the J-PARC.

References

1-17 Research on Radiation Resistance of Grease, etc. for Reprocessing Cell Equipment

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In the nuclear cycle engineering department of JAEA, an innovative centrifugal contactor system has been developed for an advanced aqueous reprocessing of FBR fuel \textsuperscript{1)}. The centrifugal contactor would be one of the key equipments to establish better reprocessing economy because of its compact size and rapid processing performance. It was already confirmed that it had a higher extraction capacity through the uranium test \textsuperscript{2}). Furthermore, in a cold experiment, the driving unit with ball bearing and grease had been operated continuously for 5000 hours that was equal to a term of an annual operation in the reprocessing plant. In this study, the equipment was operated under gamma-ray irradiation, and the durability of the driving unit was confirmed under the simulated reprocessing plant atmosphere with mechanical load, nitric acid and radiation.

The schematic diagram of experimental equipment is shown in figure 1. Generally, the extraction process consists of several centrifugal contactors, but this was one stage with a liquid self-circulation system because of the restriction of Co-60 gamma-ray irradiation facility. Two kinds of greases, RG-42R-1 and GK-1, were chosen from the viewpoint of radiation and heat resistance. The significant difference between them was that GK-1 contained MoS\textsubscript{2} as a solid lubricant to extend the lifetime. In a series of irradiation experiments, 900mL of nitric acid and 400mL of n-dodecane with the TBP concentration of 30\% were poured into it. The exposure rate (72C/kg/h) in this irradiation facility was about ten times higher than that in the reprocessing cell, so the irradiation time was set 500 hours.

After these experiments, the rate of base oil, the nitric acid concentrations and the fragment concentration in grease were measured. These measured values were not different from ones in the cold endurance experiment. These results showed that the driving units by using these greases had abilities to be operated continuously and no additive damages to ball bearings and greases was caused by combining mechanical load, nitric acid and radiation.

Furthermore the differential thermal analysis was adopted to evaluate grease degradation. Previously, the gel permeation chromatography and the infrared spectroscopy were used for the evaluations, which requested all of grease left in the ball bearing after the operation. This new method had some advantages, such as a small amount of sample and a simple pretreatment. The results of measurements are shown in figure 2. It was found that the variation of GK-1 was larger than that of RG-42R-1 in molecular weight. The reason why is as follows: the base oil of GK-1 consisted of not di-alkyl tetraphenyl but tri-alkyl tetraphenyl mainly, so that it was oxidized easily by radiation. The GR-42R-1 was more suitable than the GK-1 under the reprocessing cell atmosphere.

References

Investigation of Effect of Radiation Deterioration on Confinement Capability of Glove-box

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In the mixed-oxide (MOX) fuel fabrication facility, MOX is required to be handled in Glove-box to sustain containment of MOX into the facility. Since the Glove-box consists in some inflammable components such as plastic panel and packing, in case of fire in the facility, the static containment capability of the Glove-box may be deteriorated by pyrolysis or combustion of the components by thermal stress. Deterioration of the static confinement capability of the Glove-box may cause release of MOX to the room involving the Glove-box and to the outside of the facility and exposure of worker and public around the facility. Therefore, to confirm the safety of the facility under the fire, acquisition of fundamental data of the thermal properties of the components and establishment of quantitative evaluation method of time-course of the deterioration are very important. From this point of view, in JAEA, the thermal properties of the components have been investigated as a contract research from Japan Nuclear Energy Safety Organization (JNES)\(^1\). The purpose of this research is to examine exothermic and endothermic properties, such as starting temperatures, calorific values, temperature ranges and reaction rate data of the relative reactions of the components, as the pyrolysis properties by thermal analysis. Moreover, energy and mass release rates as source-term data for thermo-fluid analysis in the Glove-box and ventilation system and smoke generation rate as source-term data for clogging analysis of the ventilation filters with combustion of the inflammable components of the Glove-box have been also examined by using “Apparatus for Evaluation of Source-term under Fire Accident”.

Since a large amount of MOX is handled in the MOX fuel fabrication process in the actual facility, the components of the Glove-box are irradiated by radiation from MOX and radiation deterioration of the components could be caused. In this contract research, investigation of effect of the radiation deterioration on the pyrolysis properties of the components has been started from fiscal 2006. In the first place, to verify the effect, the some components of the Glove-box were irradiated under an extremely high-dose condition, 880 kGy (=10 kGy/h \(\times 88\) h), by using 6th \(^{60}\)Co cell at JAEA Takasaki. After the irradiation, the exothermic and endothermic properties and weight change as a function of temperature of the components were measured by using Simultaneous Differential Thermogravimetric Analyzer (Shimazu DTG-60H).

Figure 1 and Fig.2 show the experiment results about the effect of the radiation deterioration on the pyrolysis properties under air atmosphere condition for Polymethylmethacrylate (PMMA) and Polycarbonate (PC) as commonly used as plastic panel of the Glove-box. Initial weights of PMMA and PC were about 6 mg and 3 mg, respectively. Starting temperature of weight decrease of the irradiated PMMA was shifted to lower temperature than the non-irradiated PMMA. Moreover, endothermic peak at about 270\(^\circ\)C of the irradiated PMMA decreased than the non-irradiated PMMA and exothermic peak at about 350\(^\circ\)C appeared for the irradiated PMMA. These results mean the pyrolysis rate of the irradiated PMMA is probably faster than the non-irradiated PMMA. On the other hand, for PC, the effect of the radiation deterioration on the pyrolysis properties was not observed. In future, the more detailed investigation, for example, observation of threshold of dose affecting the pyrolysis properties, is planned.

Reference

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Corrosion Behavior of Type 316L Stainless Steel Ion-irradiated under Deformed Condition

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Irradiation assisted stress corrosion cracking (IASCC) on austenitic stainless steel (SS) is one of the problem in reactor cores. Degradation of materials under irradiation and applied stress are important to the IASCC initiation. The objective of this work is to obtain the fundamental knowledge on the synergistic effect of residual stress and irradiation on the corrosion behavior of type 316L SS. In this paper, the correlation between the corrosion behavior and microchemistry change in irradiated specimens is discussed from a viewpoint of the effect of residual stress.

In order to examine the effects of residual stress during irradiation on corrosion behavior, flat plate specimens that were plastically deformed by bending constraint and without deformation constraint were irradiated by 12 MeV Ni$^{3+}$ ion using the tandem accelerator and a vacuum chamber at JAEA- Takasaki. Plastic strain of the irradiation surface region in bent specimens was calculated to be about 2%. Irradiation temperature was 330 °C. The displacement damage levels ranged from 1 to 45 dpa. The corrosion behavior of irradiated specimens was estimated by means of single-loop electrochemical potentiokinetic reactivation (SL-EPR) testing, in which the corrosion property was evaluated using the normalized charge ($P_a$). And, three dimensional atom probe (3D-AP) analysis was conducted on the irradiated specimens to investigate the microchemical compositional change in irradiated specimens.

The dose dependence of the values of normalized charge ($P_a$) is shown in Fig.1. The $P_a$ of undeformed specimen increased rapidly with increasing dose, and saturated above 12 dpa. In case of the deformed specimen, the $P_a$ below 12 dpa was much smaller than that of the undeformed specimen. At 6 dpa irradiation, the $P_a$ for the deformed specimen was 0.01 C/cm$^2$, which was about 2 orders of magnitude lower than the value for the undeformed specimen, 5.7 C/cm$^2$. At 45 dpa irradiation, the $P_a$ was almost the same between the deformed specimen and the undeformed specimen.

Since the significant effect of residual stress on corrosion behavior was observed in the specimens irradiated to 6 dpa, microchemical composition measurement was conducted using the 3DAP. Fig.2 shows Si atom map in the deformed specimen. Inhomogeneous distribution of Si was observed. In the Si enriched region, enrichment of Ni and depletion of Cr were observed. Chemical composition was measured in seven segregation regions. The enrichment of Si composition was 4.7±0.8 at.%, and the depletion of Cr composition was 4.5±1.4 at.% in comparison with the bulk composition. In the undeformed specimen, inhomogeneous distribution solute was also observed and the segregation behavior was similar to that in the deformed specimen. Chemical composition was measured in four segregation regions. The depletion of Cr composition was 5.2±1.5 at.%, and enrichment of Si composition was 6.6±2.0 at.%. It was, therefore, speculated that the degree of Cr depletion and Si enrichment at a sink such as dislocation was greater in undeformed specimen than in deformed specimen. And, it could be suggested that radiation induced segregation behavior of solute atoms as a consequence of diffusion and annihilation of irradiation defects at sink is affected by residual stress.

The corrosion behavior of SSs was related to the diffusion behavior of the solute atoms such as Cr. It could be speculated that, due to the suppression of the segregation of solute by residual stress, deformed specimens showed higher corrosion resistance than undeformed specimens.

Reference


Fig.2 Three dimensional map of Si atoms in 316L irradiated at 330°C under the deformed condition. The volume is approximately 15 x 15 x 170 nm$^3$.

Present study is the result of “New evaluation method of material degradation considering synergetic effects of radiation damage” entrusted to Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).
1-20 Mechanical Properties of Austenitic Stainless Steel Ion-irradiated under External Stress

I. Ioka a), Y. Ishijima a), M. Futakawa b), H. Ogawa a), Y. Nakahara a) and T.Naoe c)

a)Division of Fuels and Materials Engineering, NSED, JAEA,
b)QuBS, JAEA, c)Graduate School of Ibaraki University

Irradiation creep has been studied extensively to provide data that can be used for component analysis of fusion, fast breeder and light water reactors. It is now generally recognized that the irradiation creep data were in agreement with an empirical equation 1). Moreover, there have been many irradiation experiments done under external stress to investigate stress effects on microstructural evolution. It is reported that the total nucleation rate of Frank loops was increased with increasing external stress 2). However, there are few studies on the effect of external stress on mechanical properties of the material under irradiation. The purpose of this study is to evaluate the mechanical properties of the irradiated material under external stress.

The material tested is a high chromium austenitic stainless steel (25Cr-35Ni-0.2Ti EHP) 3) which is a candidate material of cladding tube for LWR of ultra high burnup. Chemical composition of the specimen is shown in Table 1.

Table1 Chemical composition of 25Cr-35Ni-0.2Ti (wt%)

<table>
<thead>
<tr>
<th>Element</th>
<th>Fe</th>
<th>Cr</th>
<th>Ni</th>
<th>Ti</th>
<th>C</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>bal.</td>
<td>24.55</td>
<td>34.99</td>
<td>0.18</td>
<td>0.0013</td>
<td>0.0014</td>
<td></td>
</tr>
<tr>
<td>O</td>
<td>0.0011</td>
<td>0.0009</td>
<td>&lt;0.005</td>
<td>0.001</td>
<td>0.001</td>
<td></td>
</tr>
<tr>
<td>S</td>
<td>Si</td>
<td>Mn</td>
<td>P</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>L div.</td>
<td>0.0011</td>
<td>0.0009</td>
<td>&lt;0.005</td>
<td>0.001</td>
<td>0.001</td>
<td></td>
</tr>
<tr>
<td>M div.</td>
<td>0.0011</td>
<td>0.0009</td>
<td>&lt;0.005</td>
<td>0.001</td>
<td>0.001</td>
<td></td>
</tr>
<tr>
<td>S div.</td>
<td>0.0011</td>
<td>0.0009</td>
<td>&lt;0.005</td>
<td>0.001</td>
<td>0.001</td>
<td></td>
</tr>
<tr>
<td>Stress-free</td>
<td>0.0011</td>
<td>0.0009</td>
<td>&lt;0.005</td>
<td>0.001</td>
<td>0.001</td>
<td></td>
</tr>
</tbody>
</table>

The thickness of specimen is about 0.5 mm. It has three different cross-sections (S division: 1.5 mm, M division: 1.75 mm, L division: 2.0 mm in width) in order to irradiate under different stress at the same time. The surface of specimen was polished with #2400 paper.

The specimens were irradiated in triple (12 MeV Ni 3+, 1.1 MeV He + and 380 keV H + ) ion beam mode at a temperature of 300 °C using the triple ion beam facility (TIARA). The temperature of the specimen was measured by an infrared thermometer (THERMAL VISION, Nikon Co.). The displacement damage in the specimen was mainly attributed to Ni 3+ ion irradiation. The peak dose was about 10 dpa around 2 µm. The He + and H + ions were implanted in depth ranges from 1.0 to 1.5 µm using aluminum foil energy degraders. The concentrations of He + and H + ions in the implanted range were 15 appmHe and 100 appmH, respectively, which correspond to LWR condition. The dose was about 5 dpa in the implanted range of He + and H + ions.

Two specimens were irradiated. One was a stressed specimen. The other was a stress-free specimen which was prestressed at the same stress level of the stressed specimen.

The microindentation test was carried out on the surface of specimens at room temperature. A testing machine, DUH-200 (Shimadzu Co.), was used for the microindentation test. A load was applied with a loading speed of 4.4 mN/s, held 1 second and then removed. During loading, the load was continuously monitored along with the displacement with a resolution of 2 mN and 0.01 µm, respectively. The Berkovich indenter was used in the microindentation test. The obtained data were converted into the relationship between load/depth and depth (L/D-D curve), and the universal hardness was calculated from these data.

Figure 1 shows the universal hardness of irradiated region at the S, M and L divisions. The vertical axis denotes the universal hardness normalized by the unirradiated ones. The result of stress-free specimen is added to Fig.1. The

![Graph showing comparison of normalized universal hardness](image)

References
3) K.Kiuchi et al., ICAPP, June 9-13, 2002(Florida).
Evaluation of Property Change in Ultra High Purity Austenitic Stainless Steel under BWR Condition Simulated by Triple Ion Irradiation

Y. Ishijima, I. Ioka, M. Yamamoto and J. Suzuki

Division of Fuels and Material Engineering, NSED, JAEA

1. Introduction

To prevent material degradation under corrosion and LWR condition, we developed ultra high purity (UHP) grade (C + N + O < 100 wppm) and high Cr (~25wt% Cr) austenitic stainless steel. We have evaluated corrosion behavior and change of mechanical properties of UHP grade austenitic stainless steel. Decrease of carbon concentration in stainless steel decreases receptivity of irradiation assisted stress corrosion cracking (IASCC). As a result, it is considered that segregation of small amount (~10 wppm) of B and P under BWR condition causes degradation of grain boundary corrosion resistance. However, the behavior of these elements under BWR condition is not clear. The purpose of this study is to obtain basic knowledge of behavior of small amount of impurities (B and P) under irradiation.

In this paper, triple ion irradiation that simulates BWR condition was carried out for the candidate (Fe-25Cr-35Ni-0.2Ti UHP grade) and segregation of small amount of elements around grain boundary was evaluated.

2. Experimental procedures

The candidate (Fe-25Cr-35Ni-0.2Ti UHP grade) was cut to a size of 6x3x2 mm³. Irradiated area was then mechanically and electrochemically polished to a specular finish. Triple ion irradiation simulating BWR neutron irradiation condition was performed using Ni³⁺ (12 MeV), He⁺ (1.1 MeV) and H⁺ (0.38 MeV) ion beams. Aluminum film degraders were used to spread He⁺ and H⁺ irradiated area. Irradiation temperature was 573 K. Irradiation damage and ion-implantation depth were calculated using TRIM code. As a result, He⁺ and H⁺ ion-implantation depth was estimated to be 1.1~1.3 μm and irradiation damage of this area to be 75 dpa. He⁺ and H⁺ concentrations in ion-implantation area were 3 and 30 appm/dpa, respectively.

Segregation of elements around grain boundary after ion-irradiation was evaluated using Auger electron spectroscopy (AES) in which the electron beam diameter was 25 nm. After electropolish to a depth of 1.2 μm at irradiated face, evaluation of segregation was carried out at 10 nm intervals from grain boundary.

3. Results

Figure 1 shows chemical compositions of the candidate around grain boundary after ion-irradiation by AES measurement. From this result, irradiation induced segregation of Cr was observed but segregation of small amounts of impurities (B and P) were not observed by AES measurement.

At next step, 3D-AP measurement will be used to evaluate irradiation behavior and presence state of small amount of impurities.

Fig. 1 result of AES measurement
1-22 Radiation Hardening of Ferrite Phase in a Dual Phase Austenitic Steel

S. Jitsukawa a), N. Okubo a) and M Ando b)

a)NSED, b)FRDD, JAEA

Dual phase stainless steel is widely used as the structural material for LWR core component. For example, cast stainless steel for LWR components CF8 with similar chemical composition to that of 304 stainless steel often exhibits dual phase microstructure depending on the cast condition. During the initial stage of the solidification, Ni rich austenite phase is formed. This is followed by ferrite phase formation with lower Ni content. Although the Ni content is lower, this ferrite phase containing 2 to 6 % of Ni is expected to exhibit large hardening by neutron irradiation. Also, austenite phase containing lower level of Ni has lower phase stability tends to cause lower toughness after irradiation. Hardening is often accompanied by the reduction of toughness and increase in DBTT. Therefore, susceptibility to irradiation hardening of dual phase stainless steel is examined by ion irradiation method at temperatures from 200 °C to 350 °C to damage levels to 5 dpa.

An alloy with nominal chemical compositions of around 0.01C-1Si-0.7Mn-9Ni-19Cr containing 10% ferrite phase after solution annealing at 1050 °C is used. Yield stress, ultimate tensile stress and hardness at room temperature are 210 MPa, 490 MPa and 168 Hv (correspond to 2.1 GPa of average contact area pressure), respectively. Ultra micro-hardness value was about 4.5 GPa. Surface of the specimen for ion irradiation was finished by electro-polishing technique. Irradiation was carried out with 10MeV Fe ions to nominal damage levels of 0.1, 1 and 5 dpa at temperatures of 200, 280 and 350 °C. Hardness after irradiation was examined with ultra micro-hardness testing machine with indentation load of 1 g.

Hardness at twenty points was measured for one specimen after irradiation. Minimum distance between each point was about 10 micro-meter. Size of each indentation was about 1 micro-meter. Figure 1 (a) shows impressions on the irradiated surface with microstructure. Dark and light areas correspond to ferrite and austenite phases, respectively. Hardness was plotted in Fig. 1 (b). The hardness of the ferrite phase (red) was apparently higher than those in austenite regions, as shown in the figure. Damage level dependence of the hardness is plotted in figure 2. Plots for a reduced activation ferritic steel of F82H (a reference alloy) are also shown in the figure. Before irradiation, hardness for the dual phase alloy and F82H exhibited similar hardness of about 4.5 GPa. After irradiation to 0.1 dpa, some of the ferrite phase in the dual phase alloy hardened to 5.5 GPa, while small hardness change was obtained for austenite phase and F82H. At higher damage levels, hardness in most of the ferrite phase was continuously higher than austenite phase, and attained to 8 GPa at 5 dpa. Damage level dependence at 280 °C and 350 °C was qualitatively similar to that at 200 °C. Results clearly indicate that ferrite phase in the dual phase austenitic alloy is susceptible to irradiation induced hardening.

Acknowledgement

Some of the results are obtained from the research sponsored by JNES.

References


Fig. 1 Microstructure (a) and Hardness (b) after irradiation in the dual phase alloy after irradiation.

Fig. 2 Damage level dependence of hardness in the dual phase alloy. Results for F82H (a reference martensitic steel) were also shown.
Study on Irradiation Defect Generation Process and Radiation Induced Segregation in Fe-P Model Alloys

T. Tobita a), Y. Chimi a), N. Ishikawa b), S. Nakagawa c) and A. Iwase c)

a) Reactor Safety Research Unit, NSRC, JAEA, b) Advanced Science Research Center, JAEA, c) Department of Materials Science, Osaka Prefecture University

To investigate the fundamentals of irradiation embrittlement mechanisms of Reactor Pressure Vessel steel (RPVs), we have performed electron irradiation experiments using Fe-based model alloys, such as pure Fe, Fe-Cu and Fe-P. The impurities in RPVs materials have been known to have a storing effect on the irradiation embrittlement. The Cu precipitates which grow during irradiation cause irradiation hardening, while the P segregation to the grain boundary weakens the Fe-Fe bonds. Our previous study 1) shows the changes of electrical resistivity of Fe-Cu model alloys induced by electron irradiations at high temperature to evaluate the irradiation damage mechanisms, such as the behaviours of irradiation defect and Cu clustering in the materials.

In this paper, we measured the changes of electrical resistivity of several types of Fe-P model alloys induced by 2MeV electron irradiations at high temperature (523 K) to evaluate the irradiation damage mechanisms, such as the behaviours of irradiation defect and P segregation to the grain boundary in the materials. We tried to obtain the dose dependence and P content dependence of the electrical resistivity change.

The chemical composition of these materials is shown in Table 1. The Fe-P model alloys were fabricated as pure as possible to eliminate the effects of impurities. After the heat treatment at 950 °C for 10 min, the materials were quenched to keep the P atoms in supersaturated solid solution at room temperature. The specimens were cut into ribbons of 1mm width, 0.03 mm thickness, and the four electrodes were spot-welded. Figure 1 shows the schematic illustration of electron irradiation apparatus. 2 MeV electron irradiation was performed at dose rate $1.5 \times 10^{-9}$ dpa/s. The electrical resistivity was measured at 77K in LN2. In addition to these irradiation tests, thermal aging experiments at the same temperature were performed to clarify the irradiation temperature effect.

Figure 2 shows the electron dose dependence and thermal aging time dependence of the electrical resistivity change in Fe-0.1 %P and Fe-0.04 %P, where thermal aging time and electron irradiation time were almost the same. At this temperature and dose, accumulation of irradiation defects (vacancy and interstitial) is negligible 1). Therefore, the decreasing of electrical resistivity change was mainly caused by behavior of P or annihilation of thermal vacancy introduced by heat treatment.

The electrical resistivity change decreases with increasing electron dose and aging time in all materials. However, the amount of decrease is in inverse proportion to P content. And furthermore, both the amount of decreasing of electrical resistivity change decreasing induced by electron irradiation and thermal aging were almost the same. It means that detection of P segregation by electrical resistivity is impossible, or P segregation was not clearly enhanced in this electron fluence. If the decreasing of electrical resistivity change was mainly caused by decreasing of thermal vacancy introduced by heat treatment, P may obstruct the migration and annihilation of thermal vacancies.

Table 1 Chemical composition of materials (wt%)

<table>
<thead>
<tr>
<th>Material</th>
<th>Fe</th>
<th>P</th>
<th>C</th>
<th>Si</th>
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<tr>
<td>Fe-0.04%P</td>
<td>Bal.</td>
<td>0.039</td>
<td>&lt;0.0005</td>
<td>&lt;0.002</td>
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<tr>
<td>Fe-0.1%P</td>
<td>Bal.</td>
<td>0.103</td>
<td>&lt;0.0005</td>
<td>&lt;0.002</td>
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<tr>
<td>Mn S Cu O</td>
<td>&lt;0.01</td>
<td>0.0008</td>
<td>&lt;0.01</td>
<td>0.019</td>
</tr>
<tr>
<td></td>
<td>&lt;0.01</td>
<td>0.0009</td>
<td>&lt;0.01</td>
<td>0.018</td>
</tr>
</tbody>
</table>

Fig.1 Schematic illustration of electron irradiation.

Fig.2 Electron dose dependence and thermal aging time dependence of the electrical resistivity change in Fe-0.1 %P and Fe-0.04 %P.

Reference
Evaluation of Radiation Damage Created via Elastic Displacements in Oxide Ceramics

N. Ishikawa

Division of Fuels and Materials Engineering, NSED, JAEA

Radiation damage is one of the important causes of degradation of thermal conductivity and serious swelling in MOX (Mixed Oxide) fuel. Understanding of radiation damage in nuclear fuel is important for controlling fuel properties during burn up and for the prediction of fuel burn up behavior. The objectives of this study are to elucidate the irradiation parameter dominating the radiation damage of ceramics by means of accelerator experiments, and to precisely evaluate their radiation damages. Especially for simulating the process of radiation damage by elastic displacements, 10MeV Ni irradiation experiment has been performed at room temperature and at high temperature, and CeO$_2$ target which has the same crystal structure with nuclear fuel is adopted.

Thin films of CeO$_2$ were prepared on Al$_2$O$_3$ single crystal substrates, and were irradiated with 10 MeV Ni ions at room temperature and at high temperature of 400 ℃. After the irradiations, fluence dependence of change of X-ray diffraction pattern was measured in the wide fluence range of 10$^{12}$-10$^{16}$ ions/cm$^2$, in order to quantify the corresponding radiation damage. The film thickness was restricted to be about 0.3 μm which is thin enough to rule out the possibility of unrequired implantation effects.

In Fig.1 evolution of X-ray diffraction peak for different fluence is shown for CeO$_2$ irradiated with 10 MeV Ni at 400 ℃. It is found that the intensity of the peak monotonically decreases as increasing fluence, but in the high fluence region this tendency slows down. This reflects that the sample maintains its crystal structure even in the high fluence region, and the CeO$_2$ structure has high radiation resistance up to 10$^{16}$ ions/cm$^2$. It is also found that the peak position shows complex fluence dependence; it moves toward high angle side in the low fluence region (<10$^{13}$ ions/cm$^2$), and moves back to low angle side in the high fluence region (>10$^{13}$ ions/cm$^2$). This tendency is clearly demonstrated in Fig.2, where lattice parameter change decreases in the low fluence region and increases in the high fluence region. The increment of the lattice parameter for room temperature irradiation is more prominent than that for high temperature irradiation when compared at high fluence region (10$^{15}$-10$^{16}$ ions/cm$^2$), suggesting that the created oxygen defects are diminished by thermal energy in case of high temperature irradiation.

Present study is the result of “Research of highly accurate evaluation of radiation damage in advanced nuclear reactor fuel ceramics” entrusted to “Japan Atomic Energy Agency” by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).
Behavior of Advanced Materials under Heavy Irradiation

N. Okubo a), E. Wakai a), S. Yamashita a), M. Ando a), S. Otsuka a), S. Jitsukawa a), N. Hashimoto b) and S. Ohnuki b)

a) Nuclear Science and Engineering Directorate, JAEA
b) Hokkaido University

In the structural materials heavily irradiated in fusion reactor, next generation fast breeder reactor and IFMIF target etc., displacement damage and He accumulation simultaneously occur by high energy irradiation. The materials are degraded by the effect of superposition. Therefore, to understand dependences of behaviors of irradiation hardening and microstructural evolution on displacement damage and He concentration is important for modeling of irradiation behavior.

In this study, the behavior of irradiation hardening of the reduced activation ferritic steels F82H was evaluated in the high damage region. For oxide-dispersion-strengthened (ODS) martensitic steel, which was one of the candidate materials for fuel cladding used in the next generation fast breeder reactor, the microstructural evolution was conducted under irradiation.

Ion irradiation of 15 MeV-Ni\(^{4+}\) to the F82H (IEA Heat) was conducted at 270\(^{\circ}\)C in order to investigate the irradiation-hardening behavior up to 165 dpa. The change of the micro-hardness (\(\Delta H\)) measured by nanoindentation was transformed to that of yield stress (\(\Delta\sigma_y\)) by relation of \(\Delta H \approx 3 \Delta\sigma_y\). The microstructural evaluation was conducted by in-situ TEM observation under 200 keV He ion irradiation at room temperature and 550\(^{\circ}\)C for 9Cr martensitic and 12Cr ferritic ODS steels.

Dependence of irradiation hardening on the displacement damage on the F82H is shown in Fig. 1. The irradiation hardening increased gradually as increasing the damage up to 20 dpa. The slope appeared to be shelving around 40 dpa and subsequently saturating. The value of power obtained from the log-log plots of \(\Delta\sigma_y\) vs. dpa data was about 0.12 for the high dpa regime.

The vicinity of oxide particles in 12Cr ODS steels observed by in-situ TEM under He irradiation (7 appm-He) is shown in Fig. 2. The He bubble formation around the boundary between oxide particle and ferrite matrix was observed as the fluence of He ions was increased. In 9Cr ODS steels, however, the bubble formation was not observed under irradiation.

Reference
1-26 Fundamental Radiolysis Effects Relevant to Volatile Iodine Formation during Nuclear Reactor Accidents

K. Moriyama a), S. Tashiro b), Y. Maruyama a) and H. Nakamura a)

a)Thermohydraulic Safety Research Group, NSRC, JAEA
b)Fuel Cycle Facility Safety Research Group, NSRC, JAEA

Iodine is recognized as the most hazardous radioactive material in terms of public health impacts among those potentially released into the environment during a severe accident of light water reactors (LWRs). The chemical behavior of iodine compounds under irradiation in the containment vessel of LWRs during accidents was, thus, studied intensively for the last three decades1). Recently, computer programs that solve mechanistic models for the chemical kinetics of iodine compounds under LWR accident conditions were developed. To validate such models, we need fundamental radiolysis data for aqueous solutions of related materials as well as overall iodine volatility data under irradiation. We performed a series of experiments to observe changes in pure water and several kinds of aqueous solutions under gamma-irradiation for this purpose.

Table 1 summarizes the conditions of the experiments. The experiments were performed in the 7th 60Co cell at JAEA Takasaki. Each solution was irradiated in a 500 mL-separable flask while stirred with 611x30 mm glass stir bar at 510 rpm. The cover gas was either swept at a constant flow rate or kept stagnant with a vent path not to cause pressurization of the flask. The stagnant condition was set when the solution contained MEK or ammonia. After the start of the irradiation on the first day, sampling of about 40 mL of each solution was done with intervals of about 1.5 hours for four times. Then, irradiation continued through the night, and the last sampling was done just after the end of the irradiation on the next morning. Electrochemical probes (electrodes) for pH, NO3-, CO2, I-, and NH3 were used to observe changes in each solution.

Figures 1 and 2 show selected results of the analysis. Pure water showed slight decrease of pH, but NO3- was not detected though nitric acid formation was suspected. MEK solution showed significant drop of pH. In the MEK solution, CO2 was generated and reached the concentration about 10M at the end of the irradiation. CsI solution showed only slight decrease of I- concentration when pH was initially set 5 and increased slightly. While, I- almost vanished when the initial pH was 3 which did not change through the irradiation.

These data are to be used for verification of a mechanistic iodine chemistry model under development.2)

Acknowledgement
This work is a part of contract research with Japan Nuclear Energy Safety Organization (Nunes).

References
2) K. Moriyama et al., 2007 Annual Mtg. AESJ, C25 (2007) [In Japanese]

Fig. 1 pH change in irradiated solutions.

Fig. 2 Decrease of iodide ion by irradiation.

Table 1 Experimental cases and conditions

<table>
<thead>
<tr>
<th>Case</th>
<th>Solution</th>
<th>Cover gas</th>
<th>Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>1B-Air</td>
<td>Pure water</td>
<td>Air flow</td>
<td>pH/NO3^-</td>
</tr>
<tr>
<td>2-Air</td>
<td>1x10^-5M MEK</td>
<td>Air stag.</td>
<td>pH/CO3^-</td>
</tr>
<tr>
<td>2-N2</td>
<td>1 x10^-5M MEK</td>
<td>N2 stag.</td>
<td>pH/CO3^-</td>
</tr>
<tr>
<td>4A-Air (pH=5)</td>
<td>5 x10^-5M H2SO4 (pH=5)/1 x10^-5M CsI</td>
<td>Air flow</td>
<td>pH/I^-</td>
</tr>
<tr>
<td>4A-N2 (pH=5)</td>
<td>5 x10^-5M H2SO4 (pH=5)/1 x10^-5M CsI</td>
<td>N2 flow</td>
<td>pH/I^-</td>
</tr>
<tr>
<td>4A-Air (pH=3)</td>
<td>5 x10^-5M H2SO4 (pH=3)/1 x10^-5M CsI</td>
<td>Air flow</td>
<td>pH/I^-</td>
</tr>
<tr>
<td>4A-N2 (pH=3)</td>
<td>5 x10^-5M H2SO4 (pH=3)/1 x10^-5M CsI</td>
<td>N2 flow</td>
<td>pH/I^-</td>
</tr>
<tr>
<td>5-Air</td>
<td>0.1M NH3 (pH=11)</td>
<td>Air stag.</td>
<td>pH/NH3</td>
</tr>
</tbody>
</table>

MEK = methyl-ethyl-ketone
flow/stag. = flow (1L/min) or stagnant cover gas
Solution initial volume: 500mL
Temperature: 17-22°C
Dose rate, time: 1.88kGy/h, 1300min
Proton exchange membranes (PEMs) for polymer electrolyte fuel cells (PEFCs) have recently been developed by our original swift-heavy-ion based technique in addition to the γ-ray or electron-beam crosslinking and graft polymerization. The bombardment of high-energy ions produces cylindrical damage along with their trajectory in polymers. Our main focus is the use of such a localized damage area that is called a latent track. The approach using this ion track technology is being taken according to two techniques. One is the track etching and subsequent chemical modification inside the etched pores; the other is ion-track grafting, i.e., direct grafting into the activated zone. The report in the preceding issue described the former attempt including the preparation methodology and proton conductivity of the resulting membranes. We present here, as an extension of this study, their other basic characteristics as a PEM.

The commercial fluoropolymer films were employed as a matrix for maintaining a reasonable mechanical and chemical stability. A 25-μm-thick poly(vinylidene fluoride) (PVDF) film was bombarded by 3.5 MeV/n 129Xe from the TIARA cyclotron at the JAEA. According to the established procedure, track etching was then performed in a 9 M KOH aqueous solution at 80°C. The pore density ranged between 3.0×10^6 and 3.0×10^9 /cm^2, which agreed with the number of actually irradiated ions per 1 cm^2. The sulfonic acid groups were introduced into the track-etched cylindrical pores of 100 μm diameter via the γ-ray-induced styrene grafting and subsequent sulfonation.

A transmission electron microscope (TEM) was used to observe the morphology of proton-conductive, hydrophilic domains in the membranes. Figure 1 is the representative TEM micrograph of a cross section of the membrane stained by ruthenium tetroxide (RuO₄) vapor. This revealed that the stained lines with about a 100 nm width were arranged almost parallel to the through-thickness direction. Accordingly, it was confirmed that the membrane had a cylindrical electrolyte part with a diameter of about 100 nm, comparable to the size of the etched pore. In other words, we were able to prepare a novel PEM, which possessed nano-sized proton-conductive pathways extending along the ion projectile as was expected.

Figure 2 shows a plot of water uptake, defined as: 100×(W_w−W_d)/W_d, where W_w and W_d are the membrane weights in the wet and dry states, respectively, as a function of the ion exchange capacity. In all cases, the water uptake of the nano-structure controlled membranes was enhanced with an increase in the ionic content. A notable result was apparent from the comparison with the properties of not only a Nafion membrane, but also the PEMs separately obtained by the usual γ-ray-induced homogeneous grafting (result not shown). The water uptake was lower than that of the homogeneously-grafted membranes in spite of the same ion exchange capacity; for example, it decreased from 36% to 23% (up to about two-thirds) at 1.6 meq/g. An inert PVDF host without any graft electrolyte chains was found to restrict the swelling of the entire membrane. It can be concluded, therefore, that our PEM is applicable for use in a PEFC, thus extending the performance beyond the limit of conventional ones.

References
2) T. Yamaki et al., ECS Transactions 3 (2006) 103.
3) T. Yamaki et al., Electrochemistry 75 (2007) 175.
Hybrid polymer electrolyte membranes (PEM) have been widely studied for the application in intermediate temperature fuel cells\(^1\text{-}^5\). These membranes are generally prepared by incorporating inorganic particles into organic polymer electrolytes. The inorganic particles enhance the water retention and thermal stability of PEMs. However, because of the lack of covalent bonds between the two components, the inorganic particles are easily separated out from the organic PEM.

In this work, we attempted to combine the radiation grafting and silane-crosslinking to prepare a covalently crosslinked hybrid PEMs\(^1\). For this purpose, we grafted a special aromatic vinylsilane monomer, namely \(p\)-styryltrimethoxysilane (StSi), which has aromatic rings available for sulfonation and has trimethoxysilyl groups available for silane-crosslinking, into poly(ethylene-co-tetrafluoroethylene) (ETFE) base film. The StSi-grafted ETFE film was sulfonated in chlorosulfonic acid solution and then silane-crosslinked by hydrolysis and condensation in HCl solution.

![Fig. 1 Structures for the \(p\)-styryltrimethoxysilane (a) monomer and new hybrid PEM (b).](image)

Fig. 1 Structures for the \(p\)-styryltrimethoxysilane (a) monomer and new hybrid PEM (b).

In this work, we attempted to combine the radiation grafting and silane-crosslinking to prepare a covalently crosslinked hybrid PEMs\(^1\). For this purpose, we grafted a special aromatic vinylsilane monomer, namely \(p\)-styryltrimethoxysilane (StSi), which has aromatic rings available for sulfonation and has trimethoxysilyl groups available for silane-crosslinking, into poly(ethylene-co-tetrafluoroethylene) (ETFE) base film. The StSi-grafted ETFE film was sulfonated in chlorosulfonic acid solution and then silane-crosslinked by hydrolysis and condensation in HCl solution.

![Fig. 2. Effect of temperature on the proton conductivity of the new hybrid PEM and Nafion. Relative humidity, 95%.](image)

Fig. 2. Effect of temperature on the proton conductivity of the new hybrid PEM and Nafion. Relative humidity, 95%.

Figure 1 shows the structures of the monomer and the prepared new hybrid PEMs. The proton conductivity as a function of temperature is shown in Figure 2. The proton conductivity of the hybrid PEM linearly increased with increase in temperature, while for Nafion, the proton conductivity gradually increased with temperature up to 70°C, and then quickly increased in the temperature rise from 80 to 100°C. The drastic increase in conductivity of Nafion was due to the changes in its structure. Therefore, Nafion is not appropriate for application in a fuel cell operating at temperatures above 80 °C. On the contrary, in the new hybrid PEMs, no drastic increase in proton conductivity was found over the tested temperature range, indicating the stable microstructure of this membrane and its suitability for use in higher temperature fuel cells.

![Fig. 3. Thermal stability of the new hybrid PEM, Nafion, and styrene-grafted PEM (ETFE-g-PSSA).](image)

Fig. 3. Thermal stability of the new hybrid PEM, Nafion, and styrene-grafted PEM (ETFE-g-PSSA).

As seen in Figure 3, the thermal stability of the new hybrid PEMs exhibited a degradation temperature near 318°C, which was about 50 °C higher than that of the styrene-based membrane (268 °C), and was comparable to Nafion (320 °C). This enhanced thermal stability was due to the temperature tolerance of the crosslinked silicon network in the membrane. In addition, the silicon attached to the \(p\)-site aromatic rings enhanced the electronic density and thus increased the bond stability between aromatic rings and sulfonic acid groups. Furthermore, due to the silane-crosslinking structure introduced, the new hybrid membrane showed the high chemical stability than that of the divinylbenzene-crosslinked styrene-grafted PEMs.

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2-01 Utilization of Scallop Processing Waste with Graft Adsorbent

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Experimental equipment was assembled for removal of Cd\(^{2+}\) in mid-gut gland with the iminodiacetic acid adsorbents synthesized with radiation-induced graft polymerization. To check some effectiveness of the experimental equipment. The spatial distribution of the Cd\(^{2+}\) concentration in mid-gut gland in the basket and the removal rate of Cd\(^{2+}\) with regenerated adsorbents were investigated. As a result, a few variation in the Cd\(^{2+}\) concentrations of mid-gut gland was found in the basket. No significant difference of the adsorption performance was observed between regenerated adsorbents and fresh ones. These results indicate that our equipment is suitable for the purpose.

ホタテ貝の加工にともない廃棄されている中腸腺を肥料や飼料等の資源として有効利用することを目的とし、Cd\(^{2+}\)を除去するための中腸腺処理装置を開発した。この装置では、中腸腺からリンゴ酸溶液でCd\(^{2+}\)を溶出させるとともに、放射線グラフト重合法により作製した捕集材を通して溶液を循環させ、Cd\(^{2+}\)を効率良く捕集することができる。

中腸腺処理装置を用いて、中腸腺 30 kg を 0.1 M リンゴ酸溶液 150 L（固液比 0.2）で処理した。リンゴ酸溶液を 30 ℃に保ち、中腸腺を入れたカゴの下方からバブリングを行なった。捕集カラムは直径 100 mm、厚さ約 0.8 mm のイミノ二酢酸型グラフト重合捕集材 800 枚を充填したもの 1 基を使用した。リンゴ酸溶液は 3 L/min（空間速度 SV = 36 h\(^{-1}\)）の速度で循環させた。時間ごとにカゴの上面の中腸腺、溶出槽中の溶液、Cd\(^{2+}\)捕集カラムの通液直後の溶液を採取し、含有する Cd\(^{2+}\)濃度を ICP 分析装置で測定した。24 時間後については、処理のパラツキを調べるため、Fig.1 に示したカゴの 18 範所から中腸腺を採取し、Cd\(^{2+}\)残濃度を調べた。

再生したカラムを使用して Cd\(^{2+}\)除去実験を行った結果を Fig.2 に示した。中腸腺中の Cd\(^{2+}\)濃度は新品の時とほぼ同じ結果が得られた。溶出槽中の溶液、Cd\(^{2+}\)捕集カラムの通液直後の溶液についても、新品の時とほぼ同じ結果であった。捕集材の再生が正常に行われたこ
2-02 Study on Analysis of Toxic Elements in Food with Graft Adsorbent

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The demand for the analysis of toxic elements in food has increased because of the current problem of polluted food. To analyze toxic elements in food, the thiol-type adsorbent was developed in the present study. The adsorbent was prepared by radiation-induced graft polymerization of glycidyl methacrylate onto polyethylene fiber in aqueous medium and subsequently the modification with L-cystein in which active sites were blocked by zinc. When the thiol-type adsorbent was put in the cadmium solution of 10ppm, the total amount of adsorbed cadmium reached to 3.4 g/kg-adsorbent. This adsorbent is considered to have promising properties for cadmium removal.

近年、食品の有害化学物質による汚染が世界的規模で問題となったため、これまであまり問題とされなかった鉛、カドミウム、スズ、ヒ素、水銀等の有害元素の分析の要望が増えてきている。食品は不均質で、糖、蛋白質、脂質などの金属分析に対して妨害する成分が多いので、分析に際しては煩雑な前処理工程が不可欠であり、これが分析の迅速化や自動化の妨げとなっていた。

そこで我々は、金属吸着時の選択性の高い放射線グラフト重合捕集材を食品分析における金属の分離及び定量へと応用し、食品中の金属汚染の迅速化や自動化に関する検討を行ってきた。これまでの検討で、食品中の塩濃度の上昇に伴って、特にカドミウム吸着の収率低下が見られたため、より選択性の高い高分子を有する捕集材が必要としていた。

そこで本研究では、生体内において有害金属の解毒をしているメタロチオネインが、チオール基でカドミウム等を捕捉している点に着目し、新規チオール型捕集材の合成を検討した。

チオール型捕集材は、将来、食品中の有害元素除去素材にも使用するため、安全性を考慮して、チオール基を有するアミノ酸であるL-システイン（L-Cys）を官能基に用い、これとグリシジルメタクリレート（GMA）母材を結合させる合成方法を検討した。その際、a) GMAとL-Cysとをチオール基以外の官能基で結合させる方法、b) チオール型キレート樹脂は、使用直前にジスルフィド結合を還元処理してチオール基に戻して使用するが、これを還元処理なしで使用できる状態を保つ方法、の両方を満たす方法を検討した。その結果、亜鉛とL-Cysとは、中性、アルカリ性においてチオール基で結合すること1)、また、チオール基と亜鉛との反応性は、食品中に多く含まれているナトリウム、カリウム、カルシウム、マグネシウム等よりは選択性が高く、カドミウム、鉛等と比べると選択性が低い。そのため、容易に亜鉛とカドミウムの交換反応が起こると2)から、先に亜鉛とL-Cysをアルカリ条件下で反応させてチオール基を亜鉛で塞ぎ、これとGMA母材を反応させる、以下の合成方法を開発した。

ポリエチレン製不織布に対して、窒素雰囲気下で10 kGyの電子線を照射した後、グリシジルメタクリレート（GMA）及びTween20を純水と混合させて得られるエマルジョン溶液に入れて40 ℃でグラフト重合を行い、GMA母材を得た。

次に、メタノール50 mlに粒状水酸化ナトリウムを添加して1～10 ％溶液に調製した後、酢酸亜鉛0.22 gとL-Cys塩酸塩0.35 gを加えて加熱溶解し、これにGMA母材1枚（約0.1 g）を投入して、60 ℃で24時間反応させた。反応後、蒸留水で洗浄した後、真空乾燥機で乾燥させて、目的のチオール型捕集材を得た。

反応したL-Cys量の指標として、チオール基の硫黄に着目し、波長分散型蛍光X線分析装置（WDX）により測定した結果、Fig.1 に示すように、水酸化ナトリウム濃度が2 ％付近にピークを示し、最高で19 μg/cm²の硫黄濃度を示した。

このチオール型捕集材0.1 gと、10 mg/ℓのカドミウム水溶液50 mlを100 mlバイアル瓶に入れて密栓し、25 ℃の恒温水槽中で24時間良く振り混ぜた後、ICP-AESによって溶液中のカドミウムの減少量を測定し、捕集量を求めた。その結果、3.4 g-Cd/kg-adという結果が得られ、新規チオール型捕集材が、設計通りにカドミウムを捕集していることが確認できた。

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Fig.1 Effect of NaOH concentration in modification reaction on sulfur concentration in graft adsorbent.
Development of Low-loss Millimeter-wave Devices Using Fluororesin

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The development of high performance millimeter-wave planar antennas is essential for their application to satellite communications and imaging diagnostics which have become increasingly important. For fabricating such devices by using fluororesin as an antenna substrate, we propose two methods, i.e., (1) surface treatment of fluororesin substrate to increase the peel strength of copper foil, and (2) fabrication method using electro fine forming (EF2) technology.

Polytetrafluoroethylene (PTFE) films were irradiated with electron beam, and different degrees of grafting were achieved by immersing the irradiated films for different intervals in monomer solution. After the grafting, copper was attached on the surface of the films for measuring the peel strength between the metal and the grafted-films. Figure 1 shows the effect of grafting on the peel strength of the metal / grafted-PTFE. It is noted that a large increase in the peel adhesion strength was obtained by the graft polymerization on the PTFE surface. The increase in the adhesion strength between the metal and grafted films can be ascribed to cross-linking reaction or covalent bonding between the copper ion and the carboxyl group of an acrylic acid (AAc) introduced onto the thin surface layer of the PTFE films.

The dielectric constant measured by a cavity resonance method is shown in Fig. 2. The values of the dielectric constant were not significantly changed by the surface treatment. It suggests that the radiation grafting does not affect the composition of the PTFE films.

By using a micro fabrication technology EF2, ultrafine pattern such as 15 μm line width and 6μm gap width without side edges can be formed. Applying this technology for fabrication of planar-type antennas on the fluororesin substrates, we have performed an experimental production of microwave antennas having 16 elements and millimeter-wave antennas having 240 elements. It was found from the evaluation of their radiation characteristics that they have good radiation patterns. The application of the antennas to biomedical measurement system, remote monitor of heart pulse and respiration is now in progress.

In summary, PTFE films have been modified by radiation-induced graft polymerization using AAc to develop low-loss millimeter-wave antennas. As a result of the treatment, the peel adhesion strength on the PTFE was improved to be 1.64 kgf/cm. In addition, the values of dielectric constant were confirmed not to be significantly changed, which is suitable for the antenna application.

Further investigations are necessary to realize low-loss devices, such as antennas, filters and non-linear transmission lines, by using radiating grafting and EF2 fabrication technique.

References


Fig. 1 Peel strength of metal / AAc / PTFE as a function of degree of grafting.

Degree of Grafting [%] = (w2-w1) / (w1) ×100
w1 = Weight before Grafting,
w2 = Weight after Grafting

Fig. 2 Dielectric constant of the films as a function of degree of grafting.

Degree of Grafting [%] = (w2-w1) / (w1) ×100
Preparation of Ampholite-immobilized Interface by Radiation-induced Grafting

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Three types of ampholites containing dimethylamino and carboxyl groups, i.e., N(CH\(_3\))\(_2\)(CH\(_2\))\(_n\)COOH (n = 1-3), were introduced into an epoxy-group-containing polymer brush grafted onto a porous hollow-fiber membrane with a porosity of 70% and pore size of 0.36 μm. Lysozyme was captured by the amphoteric polymer brush during the permeation of a lysozyme solution buffered with a carbonate buffer (pH 9.0) across the ampholite-immobilized porous hollow-fiber membranes. The N(CH\(_3\))\(_2\)(CH\(_2\))\(_2\)COOH-immobilized porous membrane exhibited a five fold higher flux for the buffer solution and a 50-fold lower binding capacity for lysozyme at a lower molar conversion of the epoxy group into the amphoteric group, up to 17%, than the other two kinds of membranes.

1. 緒言

高分子製多孔性膜の界面に放射線グラフト重合法を用いて両性電解質を有するポリマーブラシを固定し、リゾチーム吸着容量と透過流束を両立できる吸着材料を作製し、両性電解質固定高分子鎖のコンホメーションは電荷の中和の度合いにより支配される。そこで、本研究では電荷中和の度合いを調節するために、ジメチルアミノ基とカルボキシル基との間のメチレン基数を変化させた両性電解質を多孔性膜に固定し、その性能を評価した。

2. 実験

1) PE多孔性膜への両性電解質の固定

両性電解質固定ポリマーを付与した多孔性膜の作製経路をFig. 1に示す。ポリエチレン製多孔性中空糸膜に電子線を照射後、グリシジルメタクリレート(GMA)をグラフト重合した。化学式N(CH\(_3\))\(_2\)(CH\(_2\))\(_n\)COOHによって表される両性電解質の水/1,4-ジオキサン溶液(v/v = 1/1)を用いてエポキシ基の一部を開環したのち、残存するエポキシ基をジオール基へ転化した。ここで、メチレン基の数nは1から3と変化させた。

2) 両性電解質固定膜へのタンパク質吸着

リゾチーム(Lyと略記,pI 11)溶液(pH 9.0)を両性電解質固定膜の内面から外まで一定流量で透過させた。流出液のLy濃度が供給液のそれと一致するまでLy溶液を透過させた。

3. 結果と考察

エポキシ基から両性電解質基への転化率の経時変化をFig. 2に示す。メチレン基の数n = 1および3の最終転化率は同程度で、n = 2はその約1/3であった。

nが異なる両性電解質固定膜のLy平衡吸着量と緩衝液透過流束を転化率の関数としてFig. 3に示す。n = 1および3の膜のLy平衡吸着量は転化率40%を超えると急激に増加した。Ly吸着層数(= (平衡吸着量)/(理論単層吸着量))の最大値を比較するとn = 1および3の膜は、それぞれ16および8であった。Lyを多層に吸着できたのは、荷電中和よりも荷電反発の効果の方が強いためである。一方、n = 2の膜の場合、理論単層吸着量の1/50であった。どの膜でも緩衝液透過流束の転化率依存性はなかった。ここで、n = 2の膜は、n = 1および3の膜に比べて透過流束が5倍高かった。n = 2の膜ではポリマーブリッジの伸長が抑えられ、他の膜に比べ荷電中和の効果が強いこと示された。両性電解質固定ポリマーブリッジにおいて、転化率とメチレン基の数nを変化させることによって、Ly平衡吸着量を単層吸着量の1/50から16倍まで調節することができた。

References

Hydolophilic Modification of Vulcanised Rubber by Radiation Grafting

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Rubber wiper is generally chlorinated to improve the properties such as friction, adhesion and abrasion. However, chlorination causes the degradation of wiping performance owing to the hardening of rubber surface. Thus, a novel technology of surface modification is inevitable to decrease the environmental burden of chlorination. In the present study, hydrophilic modification of vulcanised rubber by radiation grafting is evaluated as a new surface treatment for the blade rubber.

1. はじめに
加硫ゴム製品は摩擦・粘着性の低減、耐摩耗性の向上を目的として表面処理が行われている。自動車用ワイバーゴムでは、ハロゲン処理が行われている。しかし、加硫ゴム表面の親水化について検討し、摩擦低減と払拭性の双方の向上が可能なワイバーゴム表面処理技術としての可能性を評価した。

2. 実験方法
ゴム基材として、カーボンブラックを配合した厚さ2 mmの架橋天然ゴム用いた。2-ヒドロキシエチルメタクリレート(HEMA)のグラフト重合では、水を溶媒として用い、モノマー濃度30~70 wt%、照射線量10~25 kGyの範囲で電子線同時照射グラフト重合を行った。グラフト率はグラフト前後の重量増加から算出した。加硫ゴム表面に対する水の接触角は協和界面科学製接触角計により求めた。表面硬度は高分子計測器製マイクロゴム硬度計により求めた。なおウェット状態から乾燥状態への変化に伴う表面硬度変化は、水に濡らしたゴムにおいて、余剰水分を拭き取ってからの表面硬度の経時変化により確認した。摩擦の測定はピンオンディスク型摩擦試験機を用い、ガラスディスクとグラフトした架橋天然ゴムとの間の乾燥摩擦係数を測定した。

3. 結果および考察
電子線同時照射法により加硫ゴムにHEMAをグラフト重合したときの、グラフト率の照射線量依存性をFig.1に示した。照射線量の増加に伴いグラフト率は上昇し、またモノマー濃度が高いほどグラフト率が高くなった。この結果から、加硫ゴム表面へのHEMAのグラフト重合は照射線量とモノマー濃度により制御できることが分かった。次にHEMAのグラフト重合における接触角のグラフト率依存性をFig.2に示した。HEMAのグラフトに伴う接触角は70°まで低下した。これからの結果からHEMAの電子線同時照射法はオーバーゴムの表面処理として満足できる摩擦特性が得られ、さらに従来にない摩擦低減と払拭性的双方を向上するワイバーゴム表面処理技術として有望であることが分かった。

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2-06 Emulsion Graft Polymerization of Vinyl Acetate

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Grafting of vinyl acetate (VAc) on the surface of polyhydroxybutyrate (PHB) film depresses the biodegradability of PHB. After saponification of grafting layer, the graft chains of VAc were converted into biodegradable polyvinylalcohol. As a result, VAc-grafted PHB recovered intrinsic biodegradability. This grafting can lead that the PHB in use can not be degraded biologically and then its waste can be decomposed after alkaline saponification. Such characteristic of VAc-grafted PHB is applicable in the cabinet of household electrical appliances. To impart this property of off-on control of biodegradability, the grafting of VAc on PHB is inevitable. However, the grafting yield is so low that VAc solution more than 80 % is only used. It is found that emulsion of VAc in water could dramatically improve the grafting yield. In the present study, the reactivity in the emulsion grafting of VAc on PHB was compared with that in the methanol solvent.

PHB film, 150 µm thick, was prepared by thermal pressing of PHB powder (Mn=3.3×10^5). The film obtained was cut into the sample, 10mm×60mm, for grafting and then irradiated with 10kGy of electron beam in nitrogen gas. Irradiation of electron beam simultaneously induces the degradation of PHB. Molecular weight was reduced 45% of the initial value after irradiation of 10 kGy and 32 % at 50 kGy. The tensile strength decreased 10 % at 50 kGy. Hence, the irradiation dose for grafting was restricted to 10 kGy to minimize the damage of trunk PHB.

The irradiated PHB film was put into glass reactor and 150 ml of VAc emulsion was added into the same reactor. to stabilize the VAc micelles in water at 60 °C. VAc of 5 % was suspended in Nonion L-4 of the concentration from 0.2 to 1%. Instead of VAc emulsion, methanol and water were used as solvents. The reactor was maintained at 60°C to carry out the VAc grafting. The grafting reaction was evaluated by degree of grafting (Dg):

\[
D_g(\%) = \frac{W_g - W_0}{W_0} \times 100
\]

where W₀ and Wg are weight of PHB films before and after graft polymerization, respectively.

Figure 1 shows the degree of grafting in VAc emulsions, 100 % VAc and 80 % VAc in methanol. In the emulsion grafting, Dg was affected by the concentration of Nonion L-4. At 0.5 %, Dg reached the maximum value of 23 % at the grafting time of 5 h. The micelle size of VAc was minimized at 0.4 µm. In the case of 80 % VAc in methanol, Dg was only 9 % at the grafting time of 5 h. This value of Dg was two fifth of the emulsion grafting. When 100 % of VAc was used, the Dg obtained was lower than those of the emulsion graftings.

The percentage of monomer consumed in the grafting was evaluated as monomer reactivity. The monomer reactivities of the grafting in water, emulsion, and methanol listed in Table 1. Since the VAc less than 2 % can be dissolved in water, the grafting data in 2 % water were compared. Monomer reactivities were 0.62 %, 1.01 %, and 0.010 % for the grafting in water, emulsion, and methanol, respectively. In the methanol, the monomer reactivity increased with the increment of monomer concentration. At 100 % VAc the monomer reactivity became 0.046 % which is lower than that in water and emulsion of 2 % VAc. The maximum reactivity was realized at emulsion grafting. The reactivities were around 1% in both VAc concentrations of 2 and 10% in the emulsion grafting.

Table 1 Monomer reactivities of the grafting in water, emulsion, and methanol in various concentrations of monomer.

<table>
<thead>
<tr>
<th>Solvent</th>
<th>Monomer concentration [%]</th>
<th>Dg [%]</th>
<th>Monomer reactivity [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>1</td>
<td>2.6</td>
<td>0.68</td>
</tr>
<tr>
<td>Water</td>
<td>2</td>
<td>4.8</td>
<td>0.62</td>
</tr>
<tr>
<td>Water (emulsion)</td>
<td>2</td>
<td>7.8</td>
<td>1.01</td>
</tr>
<tr>
<td>Water (emulsion)</td>
<td>10</td>
<td>35.0</td>
<td>0.91</td>
</tr>
<tr>
<td>Methanol</td>
<td>2</td>
<td>0.1</td>
<td>0.010</td>
</tr>
<tr>
<td>Methanol</td>
<td>80</td>
<td>9.4</td>
<td>0.030</td>
</tr>
</tbody>
</table>

References
2-07 Adsorption Performance of Graft Amine Adsorbent for Au(III)

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To meet the demand for rare noble metals, it is necessary to reduce the amount of noble metals involved in catalysts to save the amount of such noble metals used in catalysts or increase the recycle rate of waste catalysts. Recovery of these metals from waste catalysts is carried out by a wet process which comprise of dipping waste catalysts in a strong acid to dissolve the noble metals, and separating and purifying these metals from the resulting solution by an ion exchange method using chelate resin beads or by a solvent extraction method using a chelating reagent. However, the wet process has such disadvantages that diffusion of metal ions in the chelating resin beads is a rate-determining step in the recovery from low-concentrated metal solutions; the concentration efficiency is low, etc.

The objective of this study is the preparation of a new fibrous adsorbent containing amino groups for Au(III). A combination of methacrylic acid (MAA) or N-vinylacetamide (NVA) with allylamine (AAm) was co-grafted onto the nonwoven fabric (NF) using by pre-irradiation techniques. The NF was irradiated with an electron beam up to 200 kGy in nitrogen atmosphere at ambient temperature. The irradiated NF was immersed in a monomer solution immediately deaerated by bubbling with nitrogen gas. The total concentration of the two monomers was set at 50 (wt/v) % in H₂O as a solvent, where the weight ratio of AAm/MAA or AAm/NVA in the monomer mixture ranged from 90/10 to 10/90. Co-grafting was performed at 313 K for a reaction time up to 8 hours. The degree of co-grafting (Dg) was calculated from the weight gain by before and after grafting. The resultant grafted-NF was referred to as AAm/MAA (x/y, Dg) or AAm/NVA (x/y, Dg) grafted-NF, where x/y and Dg designate the weight ratio of AAm/MAA or AAm/NVA in the monomer mixture and the degree of grafting, respectively.

Figure 1 shows the distribution coefficient of AAm/MAA and AAm/NVA adsorbents along with weight percent of AAm in the monomer. The D of AAm/NVA (50/50, 88) was 5x10⁶. This value was nine times higher than that of AAm/MAA (50/50, 100). The column mode study was conducted by using a column packed with the adsorbent named AAm/NVA (50/50, 88). Column-mode study was conducted by supplying a feeding solution of pH 3 containing 1 mM of Au(III) to the column at flow rates of 260, 520, or 1040 h⁻¹ in space velocity (SV). These breakthrough curves are shown in Fig. 2. Although the breakthrough point slightly decreased at the extremely high flow rate of 1040 h⁻¹, there are no large differences in the bed volume (BV). The flow rate of 1040 h⁻¹ is 100 times faster than that of general granular resin packed column. The breakthrough points of three flow rates were 163, 150 and 144 BV, respectively. Respective breakthrough capacities were 2.76, 2.18 and 1.95 mmol/g of Au(III) on AAm/NVA.

References
2-08 Synthesis of Graft Adsorbent with N-methyl-D-glucamine for Boron Adsorption

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Boron is widely distributed in the environment more often as boric acid or borate salt which is harmful to human organism, therefore WHO revised a drinking water guideline for boron in 1998. In response to this, a boron effluent standard was limited at 10 ppm in the water pollution control law in Japan. Then, it is expected that regulation for boron is more severe in near future. The most commonly used removal techniques for boron are chemical clarification and ion-exchange of granular resin having N-methyl-D-glucamine (NMDG). The chelating adsorbents having NMDG have been extensively investigated because NMDG has high affinity for boron adsorption\(^1\). However conventional adsorbents had a slow adsorption rate, and were difficult to adsorb a low concentration of boron efficiently. Therefore, we have been developed a new method to make more effective adsorbents for boron removal.

A non-woven polyethylene fabric as a trunk polymer purchased from Kurashiki MFG. Co. was irradiated by electron beam at voltage of 2 MeV, and current of 3 mA. Radiation induced grafting of glycidyl methacrylate (GMA) onto non-woven polyethylene fabric in aqueous medium with 0.05 wt% Tween20 as a surfactant in the presence of water was investigated. The D\(_g\) of 200 % was obtained with 5wt% GMA at 20 kGy for 60 min, which is enough for metal adsorbents. GMA-grafted fabric was chemically converted to NMDG. The maximum density 2.4 mmol/g of NMDG could be given by 5 wt% NMDG at 80 °C for 1 h.

To compare the boron adsorption performance of graft adsorbent with granular resin (NMDG density 2.2 mmol/g, particle size 0.8 mm), the column-mode adsorption was conducted. Column-mode adsorption was carried out by using the 10 ppm of boron solution at pH 7 at various flow rates. The characteristic of breakthrough was evaluated by plotting C/C\(_0\) which was the value of outflow concentration divided by the injecting concentration against bed volume (BV) as shown in Fig. 1. When the breakthrough point was determined at C/C\(_0\) of 0.05, the BV values at the breakthrough point were 235 and 138 of the graft adsorbent and the granular resin at SV10 h\(^{-1}\), respectively. Figure 2 shows the effect of flow rate on BV at the breakthrough point of the graft adsorbent and the granular resin. The BV at the breakthrough point increased with decreasing flow rate. The BV at the breakthrough point of the graft adsorbent is higher than that of the granular resin at any flow rates. At the flow rate of SV50 h\(^{-1}\), the result implied that the graft adsorbent has 4 times higher breakthrough capacity than the granular resin. The breakthrough capacity of the graft adsorbent at SV100 h\(^{-1}\) gave the same one with the granular resin at SV10 h\(^{-1}\). This result indicates that the graft adsorbent can adsorb boron 10 times faster than the granular resin.

Reference


![Fig.1 Breakthrough curve of graft adsorbent and granular resin at SV10 h\(^{-1}\)](image1)

![Fig.2 Effect of flow rate on BV at breakthrough point of graft adsorbent and granular resin](image2)
2-09 Development of Bifunctional Iminodiacetic Acid Fiber

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In this work, first, the monofunctional iminodiacetic acid fiber was prepared from polyethylene-coated polypropylene fiber (PPPE) grafted with vinylbenzylglycidylether (VBGE) according to Scheme 1. Electron (200 kGy) irradiated PPPE (0.9 denier, length 3.8 cm) was immersed in dimethyl sulfoxide solution of VBGE (50 weight %) for 24 h at 50 °C, and the weight increase of the resulting VBGE-grafted PPPE (PPPE-g-VBGE) was 91% of the weight of the trunk PPPE. PPPE-g-VBGE was functionalized by heating in ethanol solution of diethyl iminodiacetate (67 volume %) at 120 °C for 5 h using a stainless steel autoclave. After alkaline hydrolysis of ester groups, the resulting fiber FGEI was finally equilibrated with a dilute hydrochloric acid of pH 2, and was dried in vacuum. Nitrogen content and acid capacity of FGEI were 1.7 mmol/g and 4.2 meq/g, respectively.

Scheme 1 Preparation of iminodiacetic acid fiber

PPPE (PPPE-g-VBGE) was 91% of the weight of the trunk PPPE. PPPE-g-VBGE was functionalized by heating in ethanol solution of diethyl iminodiacetate (67 volume %) for 5 h at 120 °C using a stainless steel autoclave. After alkaline hydrolysis of ester groups, the resulting fiber FGEI was finally equilibrated with a dilute hydrochloric acid of pH 2, and was dried in vacuum. Nitrogen content and acid capacity of FGEI were 1.7 mmol/g and 4.2 meq/g, respectively.

FGEI exhibited excellent performances in column-mode uptake of divalent metal ions from solutions of neutral pH. For instance, breakthrough capacity of Zn(II) is as high as ca. 0.5 mmol/g at high flow rate of 1000 h⁻¹ in space velocity (SV). On the other hand, trivalent metal ions such as Fe(III) must be treated in strongly acidic solutions less than pH 2 to avoid their precipitation as hydroxides. Under such strongly acidic conditions, polymer bound iminodiacetic acid groups exist as electrically neutral or positively charged species, which interfere with uptake of trivalent cations. Then, the second purpose of this work is to resolve this problem. The introduction of sulfonate as second functional group is expected to be effective, because sulfonate exists as anionic species irrespective of pH. Negatively charged sulfonate enhances swelling of the polymer phase and also reduces the undesirable effect of positively charged species [-HN(CH₂COO⁻)]. However, the sulfonation of FGEI with chlorosulfonic acid and sulfuric acid resulted in marked weight loss of the fiber (ca. 30 %). This is probably due to cleavage of ether bonds by the strong acids. Then, we have examined the electron irradiation induced graft polymerization of sodium styrenesulfonate (SSS) onto FGEI.

FGEI-ester, FGEI in Na⁺ form, and FGEI in H⁺ form were irradiated with electron (200 kGy) and they were immersed in an aqueous solution of SSS (16.7 weight %) at 40 °C for a given time. FGEI-ester grafted with SSS was hydrolyzed and changed into the H⁺ form (FGEIS1). The other two SSS grafted fibers FGEIS2 and FGEIS3 were also changed into the H⁺ form. Results for graft polymerization of SSS are summarized in Table 1, which shows that the reactivity of SSS in the electron irradiation induced graft polymerization increases in the order FGEI-ester < FGEI(Na⁺) < FGEI(H⁺). However, too much SSS was introduced in cases of FGEIS2 and FGEIS3, and then the effect of grafted SSS on uptake of Fe(III) was examined by using FGEIS1. The results are shown in Fig. 1. The monofunctional iminodiacetic acid fiber FGEI can not effectively take up Fe(III) at pH 1.5. On the contrary, the bifunctional fiber FGEIS1 can take up Fe(III) even at high flow rate of 1000 h⁻¹ in SV. FGEI and FGEIS1 gave breakthrough capacities of 0.09 and 0.61 mmol/g for Fe(III) at the feed flow rate of SV 50 h⁻¹, respectively. Thus, the electron irradiation induced graft polymerization is useful for preparation of bifunctional chelating fibers containing sulfonate without use of harmful acids.

![Fig. 1 Column-mode uptake of Fe(III) by FGEI and FGEIS1](image-url)
Preparation of Graft Adsorbent with Ultrafine Non-Woven Fabric

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Center for Education and Training, National Nuclear Energy Agency, BATAN, JAEA

The chelating adsorbent containing phosphoric group has been extensively investigated because of its unique selectivity to the metal ion. We have prepared fibrous phosphoric chelating adsorbent (FPCA) direct synthesized by radiation induced graft polymerization. It has been demonstrated that FPCA had good ability to remove toxic metals such as Pb and Cd. This adsorbent also had 200 times higher distribution coefficient for uranium than that of a commercial adsorbent at low pH. Although the FPCA was successfully prepared for industry application, the optimum grafting condition (200 kGy dose, 20 % monomer, 10/90 wt% methanol-water, 12 h reaction time at 60°C which degree of grafting reached 250 %) need to be improved. In the present study we have made improvement by using ultrafine non-woven fabrics to get optimum condition of grafting.

Polypropylene coated polyethylene (PPPE, diameter was 15 µm) and Polyethylene non-woven fabrics (UFPE, diameter was 2 µm) were cut into rectangular shape (3 cm x 5 cm) and placed into polyethylene bag. After de-aerated with nitrogen gas, samples were irradiated with electron beam in dry ice condition. The irradiated samples were contacted with phosphoric acid monomer solution (2-hydroxyethyl methacrylate phosphoric acid (HMPA) in SDS/methanol-water) in de-aerated glass ampoule and maintained at desired reaction time and temperature in water bath. At the completion of grafting, the graft copolymer was washed by methanol.

The ability of grafting between PPPE and PE polymer is shown in Fig 1. Radiation dose used was 100 kGy, 30 % monomer, 60°C reaction temperature. The degree grafting (Dg) for UFPE reached 750 % for 6 hours and for PPPE the highest Dg was 93 %. In graft reaction, because the total surface area increased with decreasing the diameter of fiber Dg increased. The Dg in 2 % SDS solution was higher than methanol and no homo-polymerization observed. So 2 % SDS can be replaced of previous condition. Since the grafting rate of UFPE was high, grafting conditions could be optimized to 50 kGy of dose, 20 % monomer, 60 °C and 3 hours. From the FTIR spectra, the grafted samples have a very strong band at 1732 cm⁻¹, which were characteristic of carbonyl group, and the peaks at 1262 cm⁻¹ and 1151 cm⁻¹ correspond to P=O and O-P-O bonds in phosphoric group, respectively. In the SEM photos, the average diameter of UFPE of non-grafting, Dg of 59, 110, 168, 189, 299, and 479 % were 2.3, 2.6, 2.8, 3.0, 3.2, 3.4 and 3.6 µm, respectively. As a conclusion, by using UFPE the optimum grafting condition had been improved as follows; the dose was 50 kGy, 20 % monomer-2 %SDS in water solution, 60°C and 3 hours which Dg reached 230 %.

References
2-11 Synthesis of Graft Adsorbent with Amine Groups onto Polylactic Acid Nonwoven Fabric

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a)Industrial Technology Institute, Sri Lanka, b)Environment and Industrial Materials Research Division, QuBS, JAEA

1. Introduction
Graft adsorbents, which can rapidly remove and effectively recover metal ions from water resources, are attracting a lot of attention as sophisticated metal adsorbents. However, the most of them were synthesized from petroleum materials1). If plant-derived polymer, such as polylactic acid (PLA), is used as trunk polymer for graft polymerization, the consumption of petroleum can be reduced, and besides the reduction of environmental burdens is brought because of biodegradable property of themselves. In the present study, the preparation of PLA-based metal adsorbent was eagerly attempted by using radiation-induced graft polymerization.

2. Experimental
PLA nonwoven fabrics (a basis weight of 120 g/m²) were irradiated with a certain dose of electron beam in nitrogen atmosphere. The grafting was carried out by immersing the irradiated fabrics into the aqueous emulsion solution consisting of 4-chloromethylstyrene (CMS), Tween 20 (surfactant) and deionized water. After grafting, the CMS-grafted samples were further treated with 50 wt% ethylenediamine (EDA) in isopropyl alcohol (IPA) at 60°C. The degree of grafting (Dg) was estimated by the increased weight after grafting, and the amine group density was defined by the nitrogen content of EDA-modified adsorbent, respectively. The metal adsorption capacity was individually evaluated through the batch adsorption experiments. The batch adsorption tests were carried out by dipping the adsorbent of 30 mg into 50 mL of each metal ion solution of 100 ppm at 25 °C for 24 h.

3. Results and Discussion
In this work, irradiation dose of 100 kGy, CMS concentration of 3 wt%, Tween 20 concentration of 0.3 wt% and reaction temperature at 40 °C were optimum conditions for CMS grafting onto PLA. As shown in Fig. 1, the Dg of PLA nonwoven fabric can be controlled in the range of up to 115 % under the optimal graft polymerization conditions. However, the optimal graft polymerization time was determined for 4 hours because the homopolymerization of CMS was occurred after 4 hours, and then the Dg at 4 hours reached about 80 % (2.8 mmol-CMS/g-adsorbent).

In the modification process of CMS-grafted PLA with EDA, PLA was readily hydrolyzed by EDA, and its mechanical strength decreased. Therefore, to prepare PLA-based metal adsorbents with adequate performance and sufficient mechanical strength, the effect of composition of modification solvent on the introduced functional groups was investigated. As a result of trial and error, 50 wt% EDA in IPA was found to be the optimum condition, and then its EDA group density was 3.0 mmol/g-adsorbent.

The EDA-modified adsorbent showed higher affinity to Cu²⁺ and lower affinity to Cd²⁺, Pb²⁺ and Zn²⁺. As seen in Fig. 2, this adsorbent could effectively capture metal ions from weak acid and neutral solutions, and then the equilibrium adsorption amounts for Cu²⁺ at pH 5.2 and Cd²⁺, Pb²⁺ and Zn²⁺ at pH 7 were 2.5, 0.37, 0.23 and 0.21 mmol/g-adsorbent, respectively.

Reference
Preparation of New Adsorbent Containing Hydroxamic Acid Groups by Grafting for Metal Adsorption

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Introduction
Hydroxamic acid groups can form complexes with various metal ions such as Cd$^{2+}$, Fe$^{2+}$, Fe$^{3+}$, Zn$^{2+}$, Pb$^{2+}$, Ni$^{2+}$, Hg$^{2+}$, Au$^{3+}$, Ag$^{+}$, Cu$^{2+}$, Co$^{2+}$, Cr$^{3+}$, and UO$^{2+}$ 1-3). In the present work, a new adsorbent containing hydroxamic acid groups was synthesized by radiation-induced graft copolymerization of methyl acrylate (MA) onto nonwoven fabric composed of polyethylene-coated polypropylene fiber. Graft copolymerization was carried out in MA emulsion containing sodium dodecyl sulfate (SDS) as an emulsifier in the presence of water. Grafting polymerizations were studied with regard to various parameters of importance: dose, concentration of MA, reaction time, reaction temperature, and concentration of SDS. Conversion of the ester groups of the grafted copolymer into the hydroxamic groups was performed by treatment with an alkaline solution of hydroxylamine (HA).

Experiment
The trunk polymer fabrics were packed in a polyethylene bag under nitrogen atmosphere. After irradiation, fabrics were placed into a glass ampule and were evacuated. The deaerated MA solution was transferred into the ampule. The graft copolymers were reacted with hydroxylamine in a methanol-water mixture at 75 °C to convert the ester groups of grafted copolymer to hydroxamic acid groups. The percentage conversion was determined by elemental analysis.

Result and Discussion
Degree of grafting increased with increasing dose, MA concentration, reaction temperature, and reaction time and decreased with an increasing SDS concentration. The grafted copolymer produced with 2 % MA, 0.1% SDS wt/v water, dose 20 kGy, reaction time 1 h, and reaction temperature 40 °C gave degree of grafting 120 % (6.33 mmol/g of MA group) (Fig.1). The maximum percentage conversion of the ester groups of the grafted groups into the hydroxamic groups was 60 % at optimum condition: 20 % HA in methanol-water ratio (5:1), pH 13, reaction time 1 h, and 1 g of trunk polymer. The FTIR spectrum of modified copolymer shows a new characteristic absorption band of an ester at 1725 cm$^{-1}$ due to C=O stretching mode in addition to the same adsorption bands of trunk polymer as shown in Fig. 2 while the FTIR spectrum of modified copolymer shows the characteristic absorption bands of hydroxamic acid (C=O at 1711 cm$^{-1}$) and amide (N-H at 1629 cm$^{-1}$). Adsorbent containing hydroxamic acid groups can adsorb 99 % of UO$^{2+}$, 98 % of V$^{5+}$, 97 % of Pb$^{2+}$, and 96 % of Al$^{3+}$ at pH, 5, 4, 6, and 4, respectively after contacted with 100 ppb metal solution for 24 h.

References

Fig 1. Effect of reaction time on degree of grafting.

Fig 2. FTIR spectrum of (a) trunk polymer, (b) grafted copolymer, (c) adsorbent.
2-13 Preparation and Characterization of Polylactic Acid Nonwoven Fabric-based Metal Adsorbent

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1. Introduction
The graft adsorbent has been attracting attention as a sophisticated metal adsorbent because the adsorption rate is 10–100 times higher than that of a commercially available particulate resin\(^1\). However, these adsorbents are originated from petroleum resources\(^2\). If plant-derived polymer, such as polylactic acid (PLA), is used as a trunk polymer, the dependence on petroleum resource and the amount of industrial waste can be decreased to some extent. In the present study, the preparation of PLA-based metal adsorbent was attempted by using radiation-induced graft polymerization, and the metal adsorption capacity and the selectivity for various metal ions were evaluated.

2. Experimental
PLA nonwoven fabrics (fiber diameter: 18 \(\mu\)m) were irradiated with a certain dose of electron beam in nitrogen atmosphere at dry ice temperature. The graft polymerization was carried out by immersing the irradiated samples into the aqueous emulsion solution consisting of GMA, Tween 20 (surfactant) and deionized water at the desired reaction temperature. After grafting, the GMA-grafted samples were further immersed in 0.5 M of disodium iminodiacetate (IDA-Na) solution at 80 °C. The degree of grafting (Dg) was estimated by the increased weight after grafting, and the IDA group density of the adsorbent was estimated by the equilibrium adsorption capacity for \(\text{Cu}^{2+}\), respectively. The metal adsorption capacity of the adsorbent was individually evaluated through the batch adsorption experiments. The batch adsorption tests were carried out by dipping the adsorbent of 30 mg into 50 mL of each metal ion solution of 100 ppm at 25 °C for 24 h.

3. Results and Discussion
The optimum grafting conditions were basis weight of 120 g/m\(^2\), dose of 50 kGy, GMA concentration of 4 wt\%, Tween 20 concentration of 0.1 wt\%, reaction temperature at 40 °C, and the Dg can be controlled in the range of up to 200 % (4.7 mmol-GMA/g-grafted PLA) (Fig.1).

Figure 2 shows the effect of methanol (MeOH) concentration in the modification medium on the IDA group density. The IDA group density increased with the increase of the MeOH concentration. However, at the same time, the PLA was also damaged by the IDA groups themselves, and its mechanical strength was deteriorated. The IDA group density of PLA-based metal adsorbent, which had adequate metal adsorption capacity and sufficient mechanical strength, was about 500 \(\mu\)mol/g-adsorbent.

The order of metal ion selectivity of the graft adsorbent was \(\text{Cu}^{2+} > \text{Pb}^{2+} > \text{Ni}^{2+} > \text{Zn}^{2+} > \text{Cd}^{2+} > \text{Co}^{2+} > \text{Ca}^{2+} > \text{Mg}^{2+}\) and the equilibrium adsorption capacities for each metal ion were 488, 423, 418, 317, 284, 207, and 111 \(\mu\)mol/g-adsorbent, respectively. Although this selectivity was in the same order of that of a commercially available particulate resin (DIAION CR11), its adsorption capacities for heavy metal ions were within the range from 30 to 80 % of those of the DIAION CR11. This difference of the adsorption capacities was caused by the lower conversion rate, below 25 %, of the GMA-grafted chains in order to maintain the sufficient mechanical strength of the fabrics.

References
2-14 Metal Adsorption of Blend Hydrogels Based on Carboxymethyl Cellulose and Carboxymethyl Chitosan

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It is well known that hydrogels of carboxymethyl cellulose (CMC) and carboxymethyl chitosan (CMCs) are produced by gamma or electron beam irradiation with their high concentrated aqueous solution 1). The hydrogels adsorb metal ions such as Cu and Ca due to the carboxyl group and amino group on the side chain of CMC and CMCs 2). In the present report, we report metal adsorption of blend hydrogels prepared by \( \gamma \)-irradiation with the mixtures of CMC and CMCs. The adsorption of Pb and Au ions onto the blend hydrogels were controlled with the composition of the CMC and CMCs.

CMC and CMCs, which were purchased from Daicel Co. Ltd, Japan and Koto Chemicals Co. Ltd, Japan, respectively, have a degree of substitution of 0.91, and CMCs has a degree of deacetylation of 84%. The CMC/CMCs aqueous solution of 30 wt.% (blend ratio of CMC/CMCs: 100/0, 75/25, 50/50, 25/75, 0/100) were sealed in polyethylene nylon bags for air-free conditions after degassing using vacuum apparatus. The irradiations were carried out using \( {^{60}}\text{Co} \) \( \gamma \)-ray source at the Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency. The samples of paste state were irradiated to a total dose of 20 to 200 kGy (a dose rate: 10 kGy/h) at room temperature to obtain blend hydrogels. The gel fraction of the obtained hydrogels was determined gravimetrically by measuring the insoluble part after extraction of sol. The metal ion concentration in a solution was measured by Inductively Coupled Plasma Mass Spectrometer, and then the adsorption rate of Pb and Au ions with the hydrogels was calculated.

Figure 1 shows the gel fraction of CMC/CMCs blend hydrogels as a function of irradiation dose. The gel fraction increased sharply up to 30 kGy and reached a constant value in the range of 100-200 kGy. An increase in the ratio of CMC raised the gel fraction of blend hydrogel. According to gel permeation chromatography, the molecular weight of CMC and CMCs was \( 5 \times 10^5 \) and \( 6 \times 10^4 \), respectively. Therefore, crosslinking of CMC is easier to occur than that of CMCs in the same condition because of entanglement between the polymer chains, resulting in the increase in the gel fraction of the obtained hydrogels.

The nitrogen content in the blend hydrogels was determined by means of elemental analysis. The nitrogen content in the obtained hydrogels was proportional to the ratio of CMCs in the mixture, which was almost equal with the theoretical ones. This indicates that the obtained hydrogels were constructed at the initial ratio of CMC/CMCs.

The relationship between the composition of CMC/CMCs in the blend hydrogels and the adsorption rate of Pb and Au ions is shown in Figure 2. The adsorption rate of metal ions increased with increasing the composition of CMCs. The adsorption rate of Pb ions increased from 55 % to about 75-80 % in the presence of CMCts. On the other hand, in the case of Au ions, the increases in the adsorption rate appeared clearly. Although the adsorption rate with CMC/CMCs (100/0) blend hydrogel was slightly 10 %, it reached 60 % at 75/25 of ratio and increased gradually to 90 %. Therefore, it suggests that the metal adsorption with the CMC/CMCs blend hydrogels was significantly influenced by amino group on the CMCs.

References

2-15 Improvement of Physical Properties of Poly(butylene terephthalate-co-adipate) / Poly(lactic acid) Blend Film by Radiation Crosslinking

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Poly(lactic acid) (PLA) is a biodegradable polymer produced by condensation polymerization of lactic acid obtained by fermentation of starch, which resources are renewable. The variety of PLA applications include food packaging, bottles, medical and pharmaceutical products result from very favorable properties of PLA such as high transparency, strength, biodegradability, biocompatibility, and barrier properties. Nonetheless, the brittleness, low deformation at break and the lack of flexibility limit its applications. Modification of PLA by blending with biodegradable aromatic polyesters to improve its mechanical properties has not been reported. The present attempt was made to modify PLA by blending with another biodegradable aromatic flexible polymer namely poly(butylene terephthalate-co-adipate) (PBTA) shown in figure 1. Triallyl isocyanurate (TAIC) as a polyfunctional monomer has been found to be the most effective crosslinking agent for PLA to improve heat-resistance in a previous study.\textsuperscript{1,2)} The effects of electron beam (EB) irradiation in the presence of TAIC on heat-resistant and mechanical properties of the blends was investigated. The elongation at break of blend films was measured and showed PLA has improved point of hard but brittle at room temperature. As a result, elongation of crosslinked PLA with 5 phr TAIC EB-irradiated at 50 kGy was about 3 %, that crosslinked PBTA / PLA (8/2) blend with 5 phr TAIC increased approximately 300 %. Furthermore, the blend film was found to keep the shape by heating at 170 °C. The brittleness and heat-resistance of PLA were improved by the blending with PBTA and the radiation crosslinking with addition of TAIC.

References

Fig. 1 Structure of PLA, PBTA and TAIC

\begin{figure}[h]
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\includegraphics[width=\textwidth]{structure.png}
\caption{Structure of PLA, PBTA and TAIC}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{gelation.png}
\caption{Crosslinking behavior of PBTA / PLA blend films, which involve different TAIC amount, formed by EB-irradiation at 50 kGy.}
\end{figure}
Modification of Elastic Poly(lactic acid) by Radiation-induced Crosslinking

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Poly(lactic acid), PLA was irradiated using electron beams in the presence of triallyl isocyanurate at 5 phr concentration as crosslinking agent. The crosslinked PLA, of which heat resistance was improved, is applied on flexible and elastic materials by addition of plasticizer or immersion in plasticizer. The elastic PLA material has several advantages such as high holding plasticizer, heat resistance at 80 ºC and transparency. It can be concluded that crosslinking technology is beneficial to expanding application of soften PLA.

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Figure 1 Amount of plasticizer in the crosslinked PLA by EB-irradiation at 90 kGy as a function of holding time at 80 ºC.

Figure 2 TEM photograph of the crosslinked PLA with 35 % plasticizer. White area: PLA, Black area: plasticizer (PL-710)
Poly(L-lactic acid) (PLLA) is produced by condensation polymerization of lactic acid obtained by fermentation of starch, which resources are renewable. Therefore, PLLA is a typical chemically synthesized and sustainable biodegradable thermoplastic, which is expected wide usage in the near future replacing many non-biodegradable engineering plastics. The applications of PLLA include food packaging, bottles, medical and pharmaceutical products which require high thermal stability. The poly(D-lactic acid) [PDLA] and PLLA blend can co-crystallize to form stereocomplex (sc-PLA), with melting point (220 ºC) that is about 50 ºC higher than for both PLLA and PDLA. Therefore, this blend was intensively studied as a potential material with high heat stability\(^1\) and triallyl isocyanurate (TAIC) has been proved as an effective crosslinker for PLLA\(^2,3\). In this study, the properties of crosslinked sc-PLA / TAIC films by radiation with different content of TAIC and radiation doses were investigated

The melt mixture of commercial PLLA and PDLA (Mw, Mw/Mn was 9.98×10\(^4\), 1.6 and 28×10\(^4\) and 1.57, respectively) were hot-pressed at 230 ºC for 5 min and then cooled to room temperature for 5 min to form a film with thickness of 0.5 mm. The films were EB irradiated at different doses with the same dose rate of 10 kGy pass\(^-1\). Gel fractions, thermal and mechanical properties of crosslinked sc-PLA films were measured by differential scanning calorimetry (DSC), thermomechanical analyzer (TMA) and tensile measurements.

The crosslinking structures are formed in irradiated sc-PLA films with TAIC. Gel fraction of the sc-PLA films represented to the crosslinking density increased with TAIC content and radiation dose. Gel fraction seems to saturate at dose higher than 30 kGy.

DSC measurements of crosslinked samples irradiated at 50 kGy show typical crosslinking in sc-PLA mixed with more than 3 % TAIC. Neither crystallization peak nor melting peak of homo crystals (crystals of PLLA, PDLA) can be observed in DSC of sc-PLA / 3 % TAIC irradiated at dose higher than 30 kGy like non-irradiated sc-PLA film.

Figure 1 shows TMA data of the sc-PLA containing different amount of TAIC and irradiated at 50 kGy. Extension of all samples increases at 60 ºC with very small extension rate to around 200 ºC, then sc-PLA and irradiated sc-PLA films with TAIC below 1% quickly elongate and break. However, the crosslinked films containing more than 3 % TAIC withdrew the load at temperature over 250 ºC with low extension.

Figure 2 shows the stress-strain curves of sc-PLA containing 3 % TAIC irradiated at different doses. At low radiation doses, the curves of irradiated samples are similar to non-irradiated sample (curves (a)–(c)). At higher radiation doses (above 30 kGy), the mechanical properties of crosslinked samples are considerably higher than that of sc-PLA.

Our research proved that the crosslinking density increased, crosslinking network became tighter resulted in a higher young’s modulus. The crosslinked samples became harder and more stable. The combination of stereocomplex formation and radiation crosslinking caused significant improvement of heat stability of PLA material.
2-18 Novel Elastic Gel Derived from Plant-resource and Its Utilization

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Carboxymethylcellulose (CMC) gel was formed by addition of acid to CMC (CMC-acid gel). Gel fraction (weight of insoluble part/initial CMC weight) was calculated after removing uncrosslinked CMC by immersing the CMC-acid gel in water. The amount of gel increased with acid concentration. Mechanism of CMC-acid gel formation was elucidated by aggregation of CMC molecules as a result of replacement of sodium in carboxyl group with hydrogen. Gels could be prepared by three different procedures; (1) mixing of CMC and acid, (2) immersion of CMC or γ-irradiated CMC pastes in acid, and (3) γ-irradiation of CMC-acid gel. Gels with different elasticity and hardness were prepared by changing degree of substitution of CMC, molar mass of CMC, type of acid, concentrations of acid and CMC, and application of γ-irradiation.

References
2-19 Preliminary Study on the Suitability of Carboxymethylated κ-Carrageenan for Radiation Crosslinking

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Many polysaccharide derivatives have been prepared by carboxymethylation reactions in order to increase the range of potential applications of these natural polymers1). Recently, it was established that carboxymethylated polysaccharides such as carboxymethyl cellulose, carboxymethyl starch, carboxymethyl chitin and carboxymethyl chitosan, predominantly undergo crosslinking reactions when are irradiated as highly concentrated aqueous solution in paste-like state2). κ-Carrageenan is a sulfated anionic polymer consisted of alternating units of galactose and 3,6-anhydrogalactose3). κ-Carrageenan is one type of carrageenan that is capable of forming thermo-reversible gels, however it typically undergoes chain scission reactions when is exposed to high-energy radiation. This study attempts for the first time to synthesize carboxymethylated κ-carrageenan (CMkC) and determines its suitability for radiation crosslinking.

κ-Carrageenan was slurried in isopropyl alcohol / water (80/20) and alkali-activated using 40 % aqueous NaOH at 40 ºC. After 1 h, monochloroacetic acid was added and allowed to react for 3 h. Carboxymethylation was confirmed by degree of substitution (DS), Fourier transform infrared spectroscopy (FTIR) and 13C nuclear magnetic resonance analysis (NMR). For crosslinking experiments, concentrated aqueous solutions of carboxymethyl κ-carrageenan were prepared using a hybrid mixer to obtain 10-40 % (w/w) in paste-form. The samples were packed and vacuum-sealed in polyethylene / nylon bags. Gamma irradiation was conducted in 60Co facility until dose of 100 kGy, in air and ambient temperature. Crosslinking was assessed in terms of gel fraction.

DS of obtained carboxymethyl κ-carrageenan derivative measured by potentiometric back-titration was 1.20. From FTIR analysis peaks due to −COO− and −CH2− vibrations of the carboxymethyl group were observed. 13C NMR analysis revealed the occurrence of carboxymethylation in the three hydroxyl groups of κ-carrageenan disaccharide unit.

Crosslinking of carboxymethyl κ-carrageenan was successfully done and proved to be promising as shown in Fig. 1. The samples irradiated at different concentrations exhibited different degrees of crosslinking as a function of dose. Gel fractions were obtained for CMkC aqueous solution at 30-40 % concentration while no gel content was exhibited by aqueous solutions at 10-20 % concentration. The highest gel content reached 39 % at 100 kGy for the 40 % aqueous solution. Concentration effect on crosslinking dose was also evident from the graph. Increase in concentration resulted in decrease of crosslinking dose. A gel fraction of about 30% was attained at 30 kGy for 40 % of CMkC aqueous solution, while the same gel fraction at 50 kGy for 30 %. These gels were immersed in distilled water for 48 hours to estimate swelling ratio. The swelling capacity of hydrogels decreased with increase in dose, as well as with increase in polymer concentration. This can be attributed to dependence of swelling on crosslink densities of the hydrogels.

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Fig.1 Gel fraction of carboxymethyl κ-carrageenan hydrogels as a function of dose. The DS of carboxymethyl κ-carrageenan is 1.20.
Effect of Radiation Crosslinking on Stability of Bent Polyethylene Pipe

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Recently polyethylene (PE) has been widely used for water and sewage pipes because of its high strength and corrosion resistivity. Bent PE pipes, which are necessary to construct pipe lines, are usually produced by bending of straight pipes at high temperatures around the melting point of PE. When such bent pipes are used for long time, change in the bending angle takes place, which is regarded as a serious problem. In this article, the effects of radiation crosslinking on the stability of bent PE pipes are investigated.

Electron irradiation was carried out at 2MV, 2mA with a dose rate of 10 kGy/pass to induce crosslinking structure for straight high density PE pipes with 60 mm diameter (thickness 5.5 mm). The PE pipes were irradiated on both sides at doses of 30 – 100 kGy. γ-ray irradiation was done for high density PE pipes with 180 mm diameter (thickness 16.4 mm) at 80 kGy with a dose rate of 10 kGy/hr.

After irradiation, straight PE pipes heated at around the melting point were set in form to bend at the angle 90° for given time and cooled down to form bent PE pipes. The angle of fabricated PE pipes was measured with an angle meter and the appearance of the pipe surface after bending at high temperatures was also examined.

Figure 1 shows changes of the angle of PE pipes bent at 135°C after electron irradiation in a dose range of 30 – 100 kGy. For the PE pipes irradiated at 50 kGy and 80 kGy, change in the angle is small, 2 – 3°, while irradiation of PE pipe at 30 kGy and 100 kGy gives large deviations. No significant change in the surface morphology was observed between irradiated PE pipes and the original sample (before irradiation). Small angle deviations within 3° are desired in the practical application of bent PE pipes. Thus, electron irradiation at 50 – 80 kGy is preferable to fabricate stable bent pipes. As an acceleration test, the stability of bent PE pipes was examined at 60°C for 24 hrs, the results of PE pipes γ-irradiated at 80 kGy are shown in Figure 2. Irradiated pipes exhibit smaller change in the angle than that of the unirradiated one. PE pipes bent at above the melting point, 133°C after irradiation indicate small deviation of only 2°. Moreover, the appearance of the PE pipe surface after bending at high temperatures was investigated. PE pipes bent at 133°C for partial melting were rough in comparison with that bent at 126°C. From these findings, it can be concluded that crosslinking is effective to improve the stability of PE pipes bent at high temperatures around the melting point of PE.

Fig. 1 Change in the angle of PE pipes bent at 135°C after electron irradiation at 30, 50, 80 and 100 kGy.

Fig. 2 Change in the angle of γ-irradiated PE pipes when they were kept at 60°C.
The sterilization of clean rooms has been performed using a mixture of formalin/air generated by the vaporization of a commercially available formalin solution containing about 37% formaldehyde (HCHO) and 5-10% methanol (CH\(_3\)OH). The mixture in clean rooms needs to be treated after sterilization. For instance, in some food packaging factories, sterilization gas is treated by circulating through heat-catalyst reactor. It takes 2 or 3 days to complete the treatment. To improve the productivity of such factories, the development of effective treatment methods is strongly required. Electron beam is expected to be one of the alternative methods for treating organic pollutants because they can effectively be oxidized by active species (OH, O, N, O\(_3\) and so on) generated in air by electron beam irradiation. In the previous study, we estimated that formalin/air mixture (air with HCHO and CH\(_3\)OH) at the HCHO concentration of 1200 ppmv in a clean room of 2000 m\(^3\) can be treated by electron beam within about half of a day\(^1\). In this study, HCHO/air mixture (air with HCHO, without CH\(_3\)OH) is irradiated to examine the decomposition process of HCHO.

Air mixture containing 100-1060 ppmv HCHO and 1% water were irradiated with electron beams at absorbed doses of 5-80 kGys. The change in the concentration of HCHO as a function of dose is shown in Fig.1. The concentration decreased exponentially with dose. From the initial slope of the curves in Fig.1, the decrease in the HCHO concentration per 1 kGy was estimated to be 19, 35, 50, 64 ppmv / kGy for 100, 280, 480, 1060 ppmv of HCHO, respectively. These values are much larger than that of OH radical generated by electron beam, i.e., 10 ppmv / kGy. These results can be explained in terms that HCHO is decomposed by a chain reaction.

The irradiation of HCHO produced HCOOH, CO, and CO\(_2\), as shown in Fig.2. Almost all decomposition products in the irradiation of HCHO were identified. The concentrations of CO and CO\(_2\) gradually increased in a dose range of 2.5-10 kGy. On the other hand, the increase of the HCOOH concentration was suppressed above 5 kGy. It indicates that the active species like OH radicals reacted with not only HCHO but also with HCOOH. Carbon balances in the HCHO irradiation showed that 104, 93, and 95% of reacted HCHO were recovered at absorbed doses of 2.5, 5, and 10 kGy, respectively. It was found that HCOOH, CO, and CO\(_2\) were main products in the irradiation of HCHO.

Reference
Development of PCDD/Fs Treatment Technology by Gamma-ray Irradiation

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1. Introduction
The emission of dioxins to the environment was about 330 g-TEQ/yr in 2005, which was decreased by 90% compared to those in 1999. On the other hand, the amount of dioxin in analytical wastes has risen in laboratory due to a high demand on dioxin analysis. It can be much burden to store the wastes as similar to Polychlorinated biphenyl (PCB) in Japan. PCB has been treated by incineration, dechlorination, and hydrothermal oxidation etc at five facilities in Japan. However, to construct sort of facilities requires residence’s agreement and corporation, and is costly. Ionizing radiation technology is expected to be one of the economical methods for treating the dioxin wastes because we can use existing irradiation facilities without additional equipments. It can treat the wastes with a simple process. No additive or temperature control is required. Nuclear waste can also use as radiation source.

2. Experiments
Experiments on the treatment of dioxin wastes were carried out using octachlorodibenzo-p-dioxin (OCDD) (Cambridge Isotope Laboratory, Inc.) because it can be handled with lower toxicity and purchased as a crystal. The samples prepared in toluene, n-nonane, and ethanol solvents (Wako Pure Chemical Industries, Ltd.) were irradiated with gamma-ray at a dose rate of 10 kGy/h. The concentration of OCDD/F and other isomers were measured before and after irradiation using GC/HRMS (JEOL JMS-700).

3. Results and Discussion
Decomposition profile for OCDD in the three solvents is shown in Fig 1. The decomposition rate of OCDD in the three solvents increased with an absorbed dose. The highest rate was obtained from OCDD in ethanol, followed by in nonane and in toluene. This decomposition order is essentially same as G-value for the formation of solvated electron in the three solvents. This suggests that solvated electron is associated with the decomposition of PCDD. The reactions of solvated electrons with dioxins are based on the dechlorination, namely the dissociation of Cl-substituents on the aromatic rings of dioxin, leading to form lower chlorinated congeners such as hepta- and hexachlorodibenzo-p-dioxin (HpCDD and HxCDD). Thus, the decomposition behavior for chlorodibenzo-p-dioxin having from zero to eight Cl-substituents (PCDDs) was investigated and the result is shown in Fig.2. The decomposition rate for PCDDs was quit lower than that for OCDD, indicating that OCDD was dechlorinated to from lower chlorinated congeners through reactions with the solvated electrons. Although the decomposition rate for OCDD in n-nonane was higher than that for toluene, PCDD in n-nonane was less decomposed compared to PCDD in toluene. To clarify the phenomena, the dechlorination profile of OCDD was examined and found that 80 % and 20-30 % of reacted OCDD were dechlorinated in n-nonane and toluene, respectively. These values did not much change up to 160 kGy. The concentration of HpCDD formed by the dechlorination of OCDD increased in n-nonane with an absorbed dose, while reached a maximum in toluene at a dose of 50 kGy. Dechlorination is a dominant reaction for the treatment of dioxin in n-nonane. Dioxin is mainly oxidized in toluene solvent. It was found that ethanol is a good solvent for treating dioxin using an ionizing radiation.

References
2-23 Study on the Treatment of Endocrine Disrupting Chemicals in Wastewater by Ionizing Radiation

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1. Introduction
Endocrine disrupting chemicals (EDCs) in the environmental water give serious influence to aquatic animals such as mimicking the estrogen and disrupting the endocrine system in living bodies. This study is to give the validity of ionizing radiation for the treatment of trace amounts of EDCs in the environmental water. Decomposition of 17 β-estradiol (E2) and p-nonylphenols (NPs) in aqueous solution were studied using 60Co γ-ray because they have higher estrogen activities in the environmental water. Real wastewaters containing some EDCs were also irradiated. The cost for the treatment of wastewater containing EDCs by ionizing radiation was estimated based on the experimental results obtained in present studies.

2. Experimental
Real wastewater samples were obtained from secondary effluents downstream from the activate sludge system in water treatment plants. Model wastewater samples were prepared by adding E2 at 1.8 nmol dm−3 or NPs at 20 μmol dm−3 to wastewater having no estrogen activity. The samples were irradiated with γ-rays at 298 K. The concentrations of the EDCs with biological activities in the samples were measured before and after irradiation by chemical analyses and biological assays. Estrogen receptors of human and medaka were used for the yeast two hybrid assay to estimate estrogen activities, which are hereinafter defined as hEA and mEA, respectively.

3. Results and Discussion
E2 in the model wastewater was decreased as a function of dose as shown in Fig. 1. The reduction rate of E2 concentration decreased with an increase in the amount of TOC. Coexistents should influence to the decomposition of E2. The same inclination was obtained for the decomposition of NPs in the model wastewater.

The change in estrogen activity of the real wastewaters by γ-ray irradiation is shown in Fig. 2. The curves of estrogen activity have a maximum, indicating that estrogen active products were formed in the decomposition process. To eliminate the endocrine disrupting property against the human body requires an expose to the samples at about 200 Gy to reach the estrogen activity less than 1 ng dm−3. Since E2 at an estrogen activity of 500 ng dm−3 in pure water was decomposed at a dose of 50 Gy, the requirement of higher doses for the decomposition of EDCs in the real wastewaters was due to the consumption of OH radicals through reactions with coexisting organic substances having no estrogen activities.

The initial investment for electron-beam system was found to be 15 yen m−3 to treat secondary effluent in sewage treatment plants with a capacity of 10,000 m3 day−1 on the basis of 15-years operation. The consumption of electricity for an operation of the electron accelerator is estimated to be 2 yen m3. Ionizing radiation is expected to be one of the promising methods for treating EDCs in water.

Reference
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3-01 Detection of Ion-beam-induced Mutations Using Positive Selection Marker Genes in Arabidopsis

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The extensive analyses using model plant Arabidopsis showed that the ion beams have higher mutagenic effects compared to low-LET radiations\(^1\). It was also shown that the ion beams frequently induce deletion-type mutations\(^2\). Furthermore, application of the ion beams for mutation breeding demonstrated that the ion beams have wide mutation spectrum and that the ion beams can induce mutants without undesirable defects\(^3\). However, there are still unsolved points in mutagenic effects of ion beams, that is, (1) relationship between the property of ion beams and the mutation frequency or spectrum, (2) relationship between the material tissue type and the mutagenic effect, and (3) mechanism for why the recessive homozygous-like mutants, such as chlorophyll deficient mutants, were obtained in the irradiated generation. In order to clarify these points, we are analyzing the mutagenic effects of ion beams using positive selection marker genes as described below. In this year, we prepared the transgenic plant materials and determined the radiation sensitivity of them to the ion beams and gamma rays.

ribosomal protein S12 (rpsL)

The \textit{rpsL} gene that encodes ribosomal S12 protein confers streptomycin-sensitive phenotype when it is transformed into \textit{Escherichia coli}. Based on the plasmid rescue method, we can detect only the mutated genes as streptomycin-resistant colonies\(^4\). Since this method can detect large number of somatic mutations in a short time, we will analyze the mutagenic effects of ion beams with the emphasis on the difference of material tissue types.

cytosine deaminase (codA)

The \textit{codA} gene product of \textit{E.coli} deaminates 5-fluorocytosine (5-FC) into 5-fluorouracil (5-FU). Since 5-FU leads to depletion of dTTP for DNA synthesis, the plants having \textit{codA} gene can not grow on a selection medium supplemented with 5-FC\(^5\) (Fig. 1). The wild type Arabidopsis ecotype Columbia was transformed with \textit{codA} gene under the control of CaMV 35S promoter. We have selected T3 plants that are homozygous for \textit{codA} gene based on the segregation ratio on 5-FC medium. This method can be thought to detect any types of heritable mutations including large deletions.

We have determined the sensitivity of the seeds of transgenic plants for carbon ions and gamma rays (Fig. 2). The shoulder dose was 930 Gy for gamma rays, 240 Gy for 320-MeV carbon ions, and 180 Gy for 220-MeV carbon ions.

\textbf{Fig. 1} Selection of transgenic \textit{codA} plants on 5-FC-supplemented medium. T2 plants obtained by a self-pollination of transgenic plants heterozygous for \textit{codA} gene were grown on 5-FC medium. \textit{codA} plants (sensitive) and wild-type (resistant) plants were segregated in a ratio of 3 : 1.

\textbf{Fig. 2} Survival curves for the seeds of \textit{rpsL} transgenic plants exposed to ion beams and gamma rays.

\textbf{References}
1) N. Shikazono et al., Genetics 163 (2003) 1449.
2) N. Shikazono et al., Genetics 157 (2001) 379.
Mutation Induction in Orchids Using Ion Beams

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Orchid is the main export for Malaysia’s floriculture industry. Due to the increasing demand for orchid pot plants and cut flowers, new commercial varieties with new characteristics such as colour, shape and longer shelf life of flower are needed. The use of hybridization for orchid breeding is restricted by sexual incompatibility, sterility problems and long breeding time. Mutation induction by ion beam provides an alternative for improvement of orchids. In this study, we tried to produce mutants with new flower colour, shape and shelf life.

The protocorm-like bodies (PLBs) of two orchid species; Dendrobium mirbellianum and Dendrobium crumenatum were placed on 6-cm sterile petri dishes containing half strength MS (Murashige & Skoog, 1962) media. The samples covered with a Kapton film (8-µm-thick polyimide film) were irradiated with 320 MeV $^{12}$C$^{6+}$ ion beams at various doses ranging from 0 to 12.0 Gy$^1$ from the TIARA AVF cyclotron (JAEA, Takasaki). After irradiation, cultures were subcultured on to fresh ½ MS media every 4 weeks for multiplication and regeneration. The number of regenerated shoots from each dose was recorded after 2 months.

Mutation effects of carbon ions on both species were observed at tissue culture stage. In D. mirbellianum, chlorophyll mutation was detected in one of cultures irradiated at 0.4 Gy (Figure 1). The PLBs from which these plantlets originated were subcultured to obtain more of the mutants. Attempt has been made to harden one of these mutant plantlets but it could not survive the glasshouse condition. The plantlets were grown for a longer period in tissue culture environment to increase the chance of survival during hardening.

More tissue culture plantlets were transferred from time to time to the glasshouse for screening. Plantlets of D. mirbellianum are growing very slow and have not reached flowering stage.

Meanwhile, variations were also observed in some of D. crumenatum cultures. Some of the variations are shown in Figure 2. In Figure 2a, 2b and 2c are culture irradiated at 2 Gy. Figure 2a shows variations in leaf shape while elongation of stem is shown in Figure 2b. The rest of 2 Gy cultures look as a culture in Figure 2c. Slow growing culture was observed in a 0.2 Gy population (Figure 2d). The control is as shown in Figure 2e.

Differently from D. mirbellianum, D. crumenatum plants have started to flowering. The length of blooming period was recorded. This is for comparison with the control that only blooms for one day. Data are still currently collected. However, no morphological change in terms of flower colour and shape was observed.

Tissue culture seedlings were constantly transferred to the glasshouse to screen for color variation in the growing plants.

Reference

Tobacco BY-2 Cells Have a Transient and Leaky
DNA-Damage Checkpoint at G$_2$/M Phase after
Gamma-ray Irradiation

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Many types of higher plants including herbaceous plants show high radiation-tolerance at both levels of individual and cell$^{1)}$, however the tolerance mechanism is unclear in large part. By quantifying the initial yields and the repair kinetics of DNA double-strand breaks (DSBs), we found that tobacco BY-2 protoplasts can tolerate 5-times more DSBs per cell than Chinese hamster ovary CHO-K1 cells$^{2)}$ although they have a similar efficiency of DSB repair.

In this study, we analyzed cell cycle phase distribution by flow cytometry and measured micronucleus (MN) induction rate in tobacco BY-2 cells irradiated with gamma rays. As a result, the fraction of G$_2$/M-phase of the cells irradiated with 40-Gy gamma rays was more than that of the unirradiated cells at 24 h of post-irradiation culture, and the difference was disappeared at 48 h (Fig. 1). This result indicates that the DNA damage checkpoint of BY-2 cells transiently functions at G$_2$/M phase. Additionally, the percentages of the cells having at least one MN were greater in irradiated cells than in unirradiated cells (Fig. 2), suggesting that the DNA damage checkpoint of BY-2 cells is leaky. In the irradiated cells, the percentage of the cells having at least one MN peaked at 48 h after irradiation and thereafter decreased over time. This is because MN is induced through at least one cell division after irradiation, and the MN induction rate per division decreases gradually.

This unstable genome maintenance seems to be unreasonable for organism at first glance, but may be admissible for life style of higher plants: most of them must grow quickly within a limited life span for acquiring sunlight more than competitors, even if the genomes of somatic cells become unstable.

References
3-04 Effect of Ion Beam Irradiation for Microspores of Eggplant (Solanum melongena L.)

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To get thornless eggplant, microspore of eggplant ‘AE-P11’ (Solanum melongena L.) were irradiated with $^{12}$C$^{6+} \cdot 320$ MeV beams ranging from 0.5 to 20 Gy by AVF cyclotron in JAEA. The number of calli formed from irradiated microspores decreased with increasing dosage. The rate of them compared with non-irradiated treatment was 91 % at 0.5 Gy, 80 % at 1 Gy, 48 % at 2 Gy, 12 % at 5 Gy, 5 % at 10 Gy and 3 % at 20 Gy. At present about four hundred calli were formed from irradiated microspores with various doses. Regenerants from the calli will be screened for thornless.

Fig.1 Effect of $^{12}$C$^{6+} \cdot 320$ MeV ion beam on the callus formation from microspores. The number of calli was counted at 4 weeks after irradiation. The average of 3 petri dishes at one experiment were expressed as a percent of non-irradiated control.

Reference
Development of Commercial Variety of Osteospermum by a Stepwise Mutagenesis by Ion Beam Irradiation

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a) Gunma Agricultural Technology Center, b) Radiation-Applied Biology Division, QuBS, JAEA

Introduction

Osteospermum is one of the composite perennials, and characterized by a long flowering time. Recently, the production of Osteospermum has increased as potted plants and flowerbed materials. Besides the ordinary mating, bud mutation has been used for the breeding of new varieties. However, the development of new and more efficient mutagenesis techniques with an artificial manner is expected at production fields. Previously, we irradiated Osteospermum 'Mother Symphony' with ion beams, and selected 37 flower-color and morphological variants out of approximately 3,000 individuals 1). Among them, an unprecedented pastel color variant was applied to the variety registration as 'Vient Flamingo (tentative name)' in March 2007 2). Besides, many petal color variants such as white and orange could be obtained from irradiated 'Mother Symphony' that has yellow petals. Usually, Osteospermum has different colors between the upper side and under side of the petals. Interestingly, the irradiated variants included orange- and yellow-isochroous variants. However, we could not obtain a white-isochroous variant that has a potentially high market value. Therefore, in an effort to obtain a variant with white-isochroous petals, we irradiated again the white petal variant of 'Mother Symphony' with ion beams.

Materials and methods

(1) Preparation of re-irradiation material

Leaf sections (quadrilateral pieces with 0.5 to 1 cm in size) of the white variant OM7 of Osteospermum 'Mother Symphony' were put on 1/2 MS medium supplemented with 0.1 mg/L NAA and 1.0 mg/L BA. After irradiation and post-cultivation, the re-differentiated individuals were obtained.

(2) Examination of irradiation conditions

Samples covered with Kapton films were irradiated with 320 MeV carbon ion beams ($^{12}\text{C}^{5+}$) at various doses (0 to 12 Gy).

(3) Selection of variants using the optimized dose

Leaf disc sections were irradiated with carbon ions ($^{12}\text{C}^{5+}$ and $^{13}\text{C}^{6+}$) at a range of 0.1 to 5 Gy. After 24 hr, the leaf disc sections were transferred to fresh 1/2 MS medium supplemented with 0.1 mg/L NAA and 1.0 mg/L BA to induce adventitious buds. After about 1 month, grown adventitious buds were transplanted on fresh 1/2 MS medium, and cultivated to obtain plantlets. The resultant plantlets were acclimatized and then grown in a greenhouse to investigate flower color and morphological changes.

Results and Discussion

The leaf disc sections of re-irradiated variant OM7 exhibited higher sensitivity to ion beams than those of 'Mother Symphony'. The adventive bud differentiation did not occur in variant OM7 over 8 Gy of $^{12}\text{C}^{5+}$ (Fig. 1). For this reason, the optimized irradiation dose for the selection of variants from OM7 was determined to be 0.1 to 5 Gy. Up to now, several thousands of plantlets were grown, and 13 distinctive individuals were selected in terms of flower color and morphology. Among them, color variations at the upper side of petals included purple and pale orange. On the other hand, color variations at the under side of petals included pale yellow and orange. However, isochroous variant was not obtained in this experiment. Other mutations in petals and stalks included dwarf, multi-petals and petal deformations.

As mentioned above, the white petal variant OM7 was derived from irradiated 'Mother Symphony' that originally has yellow petals. Second irradiation of OM7 resulted in acquisition of variants such as purple and multi-petals that did not appear in first irradiation experiment, suggesting that re-irradiation induced a stepwise mutation in pigment synthesis and morphogenesis pathways.

Fig. 1 Sensitivity of Osteospermum to ion beams.

References

3-06 Comparison of the Mutation Inducing Effect between Ion Beams and Gamma-rays - Guineagrass and Sorghum-

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\textsuperscript{a)}Institute of Radiation Breeding (IRB), NIAS, \textsuperscript{b)}Okinawa Prefectural Livestock Research Center, \textsuperscript{c)}University of Tokyo, \textsuperscript{d)}National Institute of Floricultural Science, NAFSO, \textsuperscript{e)}BATAN, Indonesia, \textsuperscript{f)}Radiation-Applied Biology Division, QuBS, JAEA

Radio sensitivity to gamma (\(\gamma\)) ray and carbon (C) ion beam irradiation were estimated in apomictic tetraploid guineagrass (\textit{Panicum maximum} Jacq.) and diploid sexual sorghum (\textit{Sorghum bicolor} (L.) Moench.). We irradiated the seed of cv. “Natsuyutaka (NY)” guineagrass and “Zhengzu (ZZ)” and “Durra (DU)” sorghum with \(\gamma\) rays and C ion beams at varying dosages and observed the germination rate, survival rate and seed fertility of \(M_1\) plants to identify the optimum dosage of the irradiation. Gamma ray irradiation was conducted at 50, 100, 200, 300, 400, 500, 600, 800, 1000 Gy to NY seeds at the IRB, Japan and sorghum seeds in National Nuclear Energy Agency (BATAN) and compared to the control (0 Gy). The C ion beam irradiation (220 MeV) was conducted at 10, 20, 30, 40, 50, 60, 80, 100, 150, 200 Gy to NY seeds and 50, 100, 150, 200, 300 Gy to sorghum seeds at Takasaki Ion Accelerators for Advanced Radiation Application (TIARA).

The survival rate of NY by \(\gamma\) ray irradiation was not decreased up to 400 Gy but gradually decreased to near 100% lethality at 1,000 Gy (Fig. 1). The survival rate by ion beam irradiation rapidly diminished at 30 Gy with near 100% lethality at 200 Gy (Fig. 2). These data suggested that LD\(_{50}\) of NY is 600 – 800 Gy for \(\gamma\) rays and 40 – 50 Gy with C ion beam irradiation. Figure 3 illustrates the seed fertility of NY \(M_1\) plants following irradiation with C ion beams. The NY became seed sterile at 150 Gy and the dosage to induce 50 % seed sterility is estimated as 50 – 60 Gy which differs slightly from the LD\(_{50}\).

The LD\(_{50}\) of ZZ sorghum by \(\gamma\) ray irradiation is estimated as 350 -500 Gy. On the contrary, the LD\(_{50}\) of ZZ and DU by C ion beam irradiation is estimated at 30 and 50 – 60 Gy, respectively, and a difference of irradiation sensitivity is suggested (Fig.4). It is assumed that the larger sorghum seeds reduced the degree of radiation penetrance allowing some survival at 300 Gy.

We hope these data will be useful in our knock-out mutation studies focused on estimating the influence of apomixis genes in guineagrass\textsuperscript{1)} and inducing unique mutant of sorghum for bio-fuel production.

Reference


\(\begin{array}{c}
\text{Fig. 1. Survival rate of “Natsuyutaka” at varying dosages of gamma ray irradiation.}
\end{array}\)

\(\begin{array}{c}
\text{Fig. 2. Survival rate of “Natsuyutaka” at varying dosages of C ion beam irradiation.}
\end{array}\)

\(\begin{array}{c}
\text{Fig. 3. Seed sterility of “Natsuyutaka” at varying dosages of C ion beam irradiation.}
\end{array}\)

\(\begin{array}{c}
\text{Fig. 4. Survival rate of “Zhengzu” at varying dosages of C ion beam irradiation.}
\end{array}\)
3-07 The Optimum Dose of Ion Beam Irradiation for Growth of Sugarcane

F. Tohjima a), Y. Takenoshita a), T. Shirao a), T. Nagatani a), M. Ooe a), K. Ueno a), Y. Hase b) and A. Tanaka b)

a) Kagoshima Biotechnology Institute, b) Radiation-Applied Biology Division, QuBS, JAEA

Sugarcane cultivar "Ni17" has hard and harmful hair on its leaf sheath. We have intended to induce hairless or few hair mutants using ion beam irradiation to tissue cultured of leaf explants. In this study, we studied on growth of sugarcane using JAEA AVF cyclotron. The efficiency of regeneration from callus, the mutation ratio of nursery plants and the selection ratio in field were observed. We have selected 20 few hair mutants among the 5,787 regenerated plants in 2006. The optimum doses on growth were 2 ~ 3 Gy of $^{12}\text{C}^{6+}$ at 320 MeV. Furthermore, we have continued characteristic investigation and field test of the selected mutants.

Table 1 The number of tested and selected plants in 2005~2006

<table>
<thead>
<tr>
<th>Ion Energy</th>
<th>Dos (Gy)</th>
<th>No. of regenerated plants</th>
<th>No. plants at open field</th>
<th>No. of selected plants at open field</th>
<th>Selection rate (%)</th>
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<td>16</td>
<td>3</td>
<td>0</td>
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<tr>
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<td>5.78</td>
<td>188</td>
<td>20</td>
<td>0.3</td>
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</tr>
</tbody>
</table>

Fig. 1 Effect of ion beam irradiation on stem length

Fig. 2 Effect of ion beam irradiation on stem diameter

References
We worked on the ion beam breeding aimed to reduce lateral buds, or to improve flower size of Chrysanthemum cultivar 'Sanyo-ohgon'. In the individual selection of the first year, various flower shapes and days for flowering were observed in the mutation, and 45 individuals were then selected from 6,186 regenerated plants. Also, a decrease in the amount of DNA was observed in the mutants with increasing the dose of ion beam irradiation. After vegetative propagation, the line selection was performed by flowering in November or March, and two lines were then selected. Moreover, it was indicated that the re-irradiation of the ion beam was effective in additional improvement of 'Sanyo-ohgon'.

<table>
<thead>
<tr>
<th>References</th>
</tr>
</thead>
</table>
3-09  Induction of Mutations by the Ion Beam Irradiation to the Bulb-scales of Lilium cv. Acapulco.

M. Kondo a), Y. Koike a), Y. Hase b), Y. Yokota b) and H. Kobayashi a)

a) Department of Biotechnology, Niigata Agricultural Research Institute  
b) Radiation-Applied Biology Division, QuBS, JAEA

We have been working on producing marketable blue lilies. It has been impossible to produce blue lilies by crossbreeding or mutation, because they lack flavonoid 3',5'-hydroxylase (F3'5'H), which is necessary for synthesis of the blue pigment, delphinidin. Therefore, we developed an Agrobacterium-mediated transformation system for lilies to transfer the F3'5'H gene 1). For the marketing of the transformed blue lilies, we have to regulate the spreading of the pollen to prevent them from crossing with other lilies, because they are genetically modified plants. The aim of this study is to produce of male-sterile lilies. In the present study, we exposed bulb-scales of lilies to 4He²⁺ beams from the AVF cyclotron and investigated the effects on the bulb-scales.

Bulbs of the Oriental hybrid lily, Lilium cv. Acapulco, were cultured under a 16 hr light/8 hr dark cycle at 25 °C on bulb propagation medium, which consisted of modified MS medium-1/2 NH₄NO₃ and KNO₃, 40 g/l sucrose, and 3 g/l gellan gum. The diameter of the in vitro grown bulb was about 10 mm or more. The penetration range of 50 MeV 4He²⁺ was 1.45 mm. Therefore, we sliced the scales to about 1.0 mm thickness. The sliced 10 scales were placed on a regeneration medium which consisted of modified MS medium-1/2 NH₄NO₃ and KNO₃, 30 g/l sucrose, 5 g/l gellan gum, 0.1 mg/l picloram, and 0.01 mg/l 6-benzylaminopurine and covered with sterilized Kapton polyimide film (7.5 µm in thickness, 45 mm square in size, Toray-Dupon, Japan). The scales were exposed at total doses of 0 Gy – 2.0 Gy of 50 MeV 4He²⁺ beams from the AVF cyclotron.

The exposed scales were transferred to the same fresh medium and cultivated for 4 months. The culture were performed under a 16 hr light/8 hr dark cycle at 25 °C. The scales were maintained by subculturing every 2 months onto the same fresh medium under the same condition. Regenerated shoots were transferred to bulb propagation medium and cultivated for 2 months. The number of regenerated plants was counted 6 months after the ion beam exposure.

It has been suggested that the irradiation doses that suppressed the proliferation rate to 50-90% were effective in induction of mutations in cultured plant tissues 2,3). In the irradiation of 50 MeV 4He²⁺, the doses of 0.2-0.8 Gy suppressed the regeneration rate to 50-90 % of the non-irradiated control (Fig. 1). We assumed from this result that the doses of 0.2–0.8 Gy of 50 MeV 4He²⁺ were appropriate for obtaining mutants. In the irradiation of 50 MeV 4He²⁺, the doses of 0.2-0.6 Gy suppressed the proliferation rate of the calluses to 50-90 % of the non-irradiated control 4). These results suggest that scales are more insensitive to ion beams than calluses. At present, the ion beam irradiation, regeneration, acclimatization and planting to a field are in progress to select male-sterile mutants of lilies (Fig. 2).

References
Development of New Commercial Strains in Functional Mushroom by Ion Beam Irradiation

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In Pleurotus ostreatus and Lyophyllum decastes, the increase in dose causes the reduction of mycelial growth. So we examined the mutagenic effect of ion beams on Pleurotus eringii, Ganoderma lucidum and Grifora fildosa. As in Pleurotus ostreatus and Lyophyllum decastes, we found that the increase in dose causes the reduction of mycelial growth. The regeneration rate went down at 600 Gy in \textsuperscript{12}C\textsuperscript{6+} beams and few spores germinated at 700 Gy in \textsuperscript{12}C\textsuperscript{6+} beams. However, no morphological changes were observed in the fruit bodies.

References
3-11 Mutation Induction of Asiatic Hybrid Lily and Lilium × formolongi Hort. Using Ion Beam Irradiation

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a) Miyagi Prefectural Agriculture and Horticulture Research Center
b) Radiation-Applied Biology Division, QuBS, JAEA

We investigated mutants of Asiatic hybrid lily and Lilium × formolongi Hort. induced by ion beam irradiation. Six flower color mutants were observed in M₁ generation of Asiatic hybrid lily, and Two pollenless mutants were observed in M₂ generation of L × formolongi. Carbon ion irradiation seemed to be more effective than Helium ion to induce mutants of Asiatic hybrid lily.

1. はじめに

スカシユリ (Asiatic hybrid lily) は、赤、桃、橙、黄等の花色で球根により繁殖し、シンテッポウユリ (Lilium × formolongi Hort.) は白花で種子繁殖性の園芸植物である。本試験では品種改良を目的に、スカシユリとシンテッポウユリに対して、イオンビームによる突然変異を誘発し、花の諸形質における遺伝的変異の拡大をねらった。

2. 実験方法

無菌培養で増殖したスカシユリの2品種ロリーポップ（花色：赤白）とチャンティ（花色：桃）のりん片に炭素12C6+(320 MeV) またはヘリウム4He2+(100 MeV)を照射した。その後にりん片から再生した個体をM₁世代とし、ほ場で栽培し開花時に変異の有無を調査した1)。シンテッポウユリについては、品種オーガスタの種子に炭素12C6+(320 MeV)を照射処理しM₁世代を育成した。M₁世代の自殖により変異した遺伝子座をホモ化し、得られたM₂世代をほ場で栽培し開花時に変異の有無を調査した。

3. 結果及び考察

スカシユリでは花色に関する6系統の変異体が得られた。ロリーポップの変異体4系統は全てFig. 1(B)に示した花の白色化であった。チャンティではFig. 1(C)に示した花色の変異体と、花被片の先端が白い変異体が得られた。ロリーポップにおいて、炭素イオンとヘリウムイオンの効果を同線量で比較した場合、突然変異率は、炭素イオンが高い傾向であった（Table 1）。シンテッポウユリのM₂世代では、変異体が観察されなかった。M₂世代では花色等の変異は無かったが、花粉が生産されない2系統の変異体を得ることができた（Table 2）。

Table 1  Number of observed mutants and estimated mutation rates in Asiatic hybrid lily by ion beam irradiation.

<table>
<thead>
<tr>
<th>Cultivar</th>
<th>Ion</th>
<th>Dose (Gy)</th>
<th>No. of M₁</th>
<th>No. of Mutants</th>
<th>Mutation Rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lolly pop</td>
<td>12C⁶⁺</td>
<td>0.5</td>
<td>90</td>
<td>1</td>
<td>1.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.0</td>
<td>40</td>
<td>1</td>
<td>2.5</td>
</tr>
<tr>
<td></td>
<td>4He²⁺</td>
<td>0.5</td>
<td>336</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.0</td>
<td>232</td>
<td>2</td>
<td>0.86</td>
</tr>
<tr>
<td>Chanty</td>
<td>12C⁶⁺</td>
<td>0.5</td>
<td>75</td>
<td>2</td>
<td>2.7</td>
</tr>
</tbody>
</table>

Table 2  Number of observed mutants and estimated mutation rates in L × formolongi by carbon ion irradiation.

<table>
<thead>
<tr>
<th>Cultivar</th>
<th>Ion</th>
<th>Dose (Gy)</th>
<th>No. of M₁</th>
<th>No. of Mutants</th>
<th>Mutation Rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Augusta</td>
<td>12C⁶⁺</td>
<td>1</td>
<td>445</td>
<td>1</td>
<td>0.22</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2</td>
<td>521</td>
<td>1</td>
<td>0.19</td>
</tr>
</tbody>
</table>

Fig. 1  Mutation of flower color in Asiatic hybrid lily. Normal (A) and Mutant (B) of cultivar "Lolly pop". Normal (C) and Mutant (D) of cultivar "Chanty".

Reference
3-12 Dose Response and Mutation Induction by Ion Beam Irradiation in Chrysanthemum

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a)Kyoto Prefectural Institute of Agricultural Biotechnology, b)Radiation-Applied Biology Division, QuBS, JAEA

Recently irradiation of ion beam has become a new method for mutation breeding of chrysanthemum. This study was conducted to investigate the effect of ion beam irradiation on mutant induction of chrysanthemum cultivars (‘H13’ and ‘Shiroyamate’). Irradiation of ion beam induced flower colour changing; that is, yellow mutant was successfully obtained from ‘Shiroyamate’, and vermilion and white/red mutants were also induced from ‘H13’. These results suggest that the ion beam irradiation would be valuable for mutation induction in chrysanthemum flower.

盆栽クギクは、仏花として出荷期が決まっているが、天候による開花変動が大きく、産地では危険分散のため多くの品種が作付けられている。こうした中、京都府では開花変動の少ない赤色系統‘H13’（赤紫色）を育成した。しかし、盆栽クギクとしては赤・白・黄の3色セットが望まれる。そこで、イオンビームを用いた突然変異育種法に取り組むこととした。イオンビームは、ガンマ線やX線などの放射線を用いた育種と比較して幅広い変異を示すことが知られており1)、キクでは花色変異品種をはじめ、多くの品種が育成されている。

本研究では、‘H13’及び在来白色品種‘白山手’から、イオンビーム照射と培養により白色や黄色等に変化した品種を育成するために、花弁あるいは葉片にイオンビームを照射し、変異体の作出を試みた。

‘白山手’の花弁を3〜5 mmに調製し、NAA 10 mg/lとBA 3 mg/lを添加したMS培地を10ml含む60 mmシャレに置床した。一方、‘H13’は葉片を3〜5 mmに調製し、NAA 0.1 mg/lとBA 0.5 mg/lを添加したMS培地を10ml含む60 mmシャレに置床した。次に、日本原子力研究開発機構（高崎市）イオン照射研究施設のAVFサイコトロンを用いて、これらサンプルに加速粒子^{12}C^{5+}、エネルギー220 MeVで0〜32 Gyの範囲で照射し、培養によりシュートを再生させた。順次、順化・育苗した後、‘白山手’はビニールハウスに定植、H13は4号ポットに鉢上げしてガラス温室内で栽培し、開花調査を実施した。

‘白山手’では、イオンビームの線量が高くなるに伴い、黄色花及び無花粉株の出現率が高くなった（Table 1 and Fig. 1）。また、葉の波立ちや矮化などの変異率も高まり、8 Gy以上では半数以上の個体に変異が現れた。これら結果から、‘白山手’の花弁に照射するイオンビームの線量は1 Gy以下が適すると考えられる。

‘H13’への照射では、濃赤色、朱色、桃色などの花色変異体が得られた（Fig. 1）。また花形や花房にも変異が認められた。現在、これら再生植物体を温室に定植し、慣行法により栽培を行っている。今後、花色や草型等の特性を調べ、有望系統を選択するとともに、得られた花色変異株に再度イオンビームを照射し、さらに花色変異の幅を広げていきたいと考えている。

Table 1 Mutation induction in ‘Shiroyamate’ by ^{12}C^{5+} ion beam irradiation

<table>
<thead>
<tr>
<th>Does (Gy)</th>
<th>No. of plants</th>
<th>Flower mutation (%)</th>
<th>Pollen sterility (%)</th>
<th>Leaf mutation (%)</th>
<th>Dwarf (Gy)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cont</td>
<td>30</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0</td>
<td>126</td>
<td>0.8</td>
<td>4</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1</td>
<td>482</td>
<td>2.3</td>
<td>1</td>
<td>6.8</td>
<td>0.8</td>
</tr>
<tr>
<td>2</td>
<td>424</td>
<td>4</td>
<td>3.8</td>
<td>10.1</td>
<td>5.4</td>
</tr>
<tr>
<td>3</td>
<td>443</td>
<td>7.4</td>
<td>2.7</td>
<td>23.3</td>
<td>12.2</td>
</tr>
<tr>
<td>4</td>
<td>360</td>
<td>9.7</td>
<td>11.1</td>
<td>28.9</td>
<td>7.5</td>
</tr>
<tr>
<td>16</td>
<td>88</td>
<td>1.1</td>
<td>0</td>
<td>10.2</td>
<td>0</td>
</tr>
<tr>
<td>32</td>
<td>46</td>
<td>6.5</td>
<td>0</td>
<td>8.7</td>
<td>0</td>
</tr>
</tbody>
</table>

*Flower mutation indicates yellow mutants
Leaf mutation indicates that leaf shape was mutated from serrated to round

Fig. 1 Selected mutants from ‘Shiroyamate’ and ‘H13’
A: ‘Shiroyamate’ (Normal)
B: Yellow variety from ‘Shiroyamate’
C: ‘H13’ (Normal)
D: Vermilion variety from ‘H13’
E: Red/white variety form ‘H13’
F: Cylinder-shaped variety from ‘H13’

Reference
3-13 Induction of Thornless Yuzu Mutant by Heavy Ion Beam Irradiation

Y. Matsuoa), Y. Haseb), Y. Yokotab), I. Narumib) and E. Ohyabua)

a) Saga Prefectural Agricultural Research Center, b) Radiation-Applied Biology Division, QuBS, JAEA

Yuzu (Citrus junos) tree bears long thorns that deteriorate fruit quality. We are trying to produce thornless mutant of Yuzu by carbon-ion irradiation. Cut surface of lower hypocotyls were exposed to carbon ions with a total energy of 320 MeV. Based on the shoot regeneration rate, we determined less than 8 Gy is appropriate for our culture system. We successfully obtained completely thornless mutant together with several mutant plants with weak thorns or lesser number of thorns. We grafted these mutant plants into rough lemon (Citrus jambhiri) to grow up quickly, and will examine their thornless traits.

ユズは樹体に長いトゲが発生するため、栽培管理上障害となっている。さらに、トゲによる刺傷はユズ果や腐敗果の発生など果実の品質性を低下させる。そこで、ユズ実生の胚軸断面からシュートを再生させる培養系に重イオンビームを照射しトゲ消失変異体の作成を試みた。今回はシュート再生率などを指標とした照射条件や変異体作成に最適な線量の検討を行うとともに、再生シュートを育成し、トゲ消失変異個体を得たので報告する。

試験1 ユズ (Citrus junos Sieb.ex Tanaka)種子を1/2MS培地に播種後、3週間目に胚軸を2cm程度残し切断した。さらに、1週間後に切断面より発生したカルスに重イオンビームを照射した。照射線種はCイオン (12C6+ 総エネルギー320 MeV)を用い、照射線量は1 Gy、2 Gy、4 Gy、8 Gy、16 Gy、32 Gy、64Gy、128Gy、256Gyの9線量区とした。照射施設は原子力機構高崎量子応用研究所のAVFサイクロトロンを用いた。照射後、再分化個体数及び根の伸長量を調査した。

試験2 試験1の結果を踏まえて、線量を1 Gy、2 Gy、4 Gy、8 Gy、16 Gy、32 Gy、64Gy、128Gy、256Gyの4線量区で照射を実施し、再分化を確認後、解剖培地へ継代培養した。さらに、培土に鉢上げし、トゲの発生が見られない個体についてはラフレモン等に寄せ接ぎを行い伸長促進を図りトゲの発生状況を確認した。

試験1 2週間後の再分化シュートの発生率は、2 Gy以下では100%以上であり、また8 Gyでも60%は再分化した。なお、シュートの発生率が100%を越えるのは1胚軸より複数発生したものである。照射線量が32 Gy以上の場合シュート発生は認められなかったが、根の伸長は確認できた。根の伸長長については、照射線量が増加しても著しい影響は見られなかった。これは、重イオンではX線等とは異なり、到達深度が浅いため、根の発生阻害が起こることに考えられ、照射時には植物体根部（地下部）の保護は必要ないと考えられる。以上の結果より、ユズ実生に重イオンビームを照射する線量は8 Gy以下で検討していくこととした。

試験2 各線量区とも200個体前後に照射を実施した結果、2 Gy区で完全無刺個体を獲得することができた。また、トゲの発生がほとんど見られない個体は1 Gyを除き各線量とも出現した。これらの個体以外に、初期生育が緩慢で判断がつかない個体もあったため、ラフレモン等に寄せ接ぎを実施し確認を急ぎたい。

Table 1. Shoot regeneration rate from hypocotyls irradiated with carbon ions.

<table>
<thead>
<tr>
<th>Dose (Gy)</th>
<th>No. of shoots 1 week after irradiation</th>
<th>Regeneration rate (%)</th>
<th>No. of shoots 2 week after irradiation</th>
<th>Regeneration rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>10</td>
<td>11</td>
<td>110</td>
<td>15</td>
</tr>
<tr>
<td>1</td>
<td>10</td>
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<td>100</td>
<td>12</td>
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<td>11</td>
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<td>4</td>
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<td>32</td>
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<td>0</td>
</tr>
<tr>
<td>64</td>
<td>10</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>128</td>
<td>10</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>256</td>
<td>10</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 2. Effects of carbon ion irradiation on root growth.

<table>
<thead>
<tr>
<th>Dose (Gy)</th>
<th>No. of roots 3 week after irradiation</th>
<th>Root length (mm)</th>
<th>No. of roots 4 week after irradiation</th>
<th>Root length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
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<td>8</td>
<td>12.3</td>
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</tr>
<tr>
<td>1</td>
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<td>7</td>
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<td>256</td>
<td>10</td>
<td>10</td>
<td>10.4</td>
<td>4</td>
</tr>
</tbody>
</table>

Figure 1. (a) Thornless Yuzu mutant. (b) Normal shoot with long thorns that is regenerated from the same hypocotyls.
Producing New Gene Resources in Fig by Using Ion-beam Irradiation

I. Asami a), S. Fukuta a), S. Kuroyanagi a), T. Ooya a), Y. Hase b), Y. Yokota b) and I. Narumi b)

a) Aichi-ken Agricultural Research Center, b) Radiation-Applied Biology Division, QuBS, JAEA

The goals of our study are to produce new gene resources in fig by mutation induction with ion-beam irradiation, because the breeding by varietal crossing in fig is very difficult. As the first paper of our study, we report the influences of carbon ion beams and soft X-rays irradiation to in vitro lateral buds on mutation induction in fig 'Masui-dorfin'. In vitro buds were irradiated with carbon ion beams (320 MeV $^{12}$C $^{6+}$) at various doses of 0 – 40 Gy at TIARA, and with soft X-rays (100 KVp, 5 mA, 191 Gy/Hr) at Aichi Agric.Res.Ctr. at various doses of 0 – 160 Gy. The efficiency of survival rate of buds, the fresh weight and the length of lateral shoots, and the rate of shoot that grows over 1 cm were observed. The optimum doses for carbon ion beams and soft X-rays were 5 Gy of 320 MeV $^{12}$C $^{6+}$ and 60 Gy of soft X-rays, respectively.

Fig.1 Influence of 320 MeV carbon ion beam irradiation to on growth of in vitro lateral buds in Fig 'Masui-dorfin'.

Fig.2 Influence of soft X-ray irradiation on growth of in vitro lateral buds in Fig 'Masui-dorfin'.

Reference
1) 平田尚美. 農業技術体系−果樹編5. 東京. 農文協. イチジク, 基29 (1993).
3-15 Induction of New Color Variation by Irradiation of Ion Beams to Light Yellow ‘Jinba’

T. Toyoda a), H. Watanabe b), K. Emoto a), S. Yoshimatsu b), Y. Hase c) and S. Kamisoyama a)

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‘Jinba’ is a white chrysanthemum. In 2002, we found a light yellow variant of ‘Jinba’ at south area of Oita prefecture. So, we have intended to induce deep yellow ‘Jinba’ using ion beam irradiation(12C6+:320MeV, 1-2-3 Gy) to this light yellow ‘Jinba’. Leaves or petals were cut and cultured on the regeneration MS medium and irradiated. Five mutants (2 Gy:2 mutants, 3 Gy:3 mutants) which showed deep yellow at flower bud time were obtained. But their petals gradually became white toward flowering.

秋輪ギク「神馬」(白)は、大分県南部の佐伯市を中心
に多く栽培されている。しかし、「神馬」の栽培特性に
類似した高品質の黄色品種が多く、黄色品種を多く生
産できないことが、生産・販売戦略上大きな障害とな
っている。また、花き研究所では、平成14年度に佐伯
市蒲江町で発見された「神馬」淡黄色系統について、
系統選抜を行い、濃黄色系統を選抜してきたが、開花
時に退色する欠点を改善されていない。

そこで、「神馬」淡黄色系統に対して花色変異に有効
なイオンビーム照射を行い、花色変異の誘発を試みた。

供試材料は、「神馬」淡黄色系統（以下元株）無菌培
養中の葉片及び花弁を用いた。葉片は2×5mm、花弁は
2×5～7mm 程度に切断し、不定芽形成培地（MS培地
+NAA2.0mg/L+BA1.0mg/L（ショ糖3%、寒天
0.8%、pH5.8））に植床し、1週間程度培養したものに、
1〜2月21日にイオンビームを照射した。照射後、同じ不定芽形
成培地に継代し、3〜4週間培養後、不定芽伸長培地（MS培地
+NAA0.01mg/L+BA0.05mg/L（ショ糖3%、寒天
0.6%、pH5.8））へ移植した。照射は、TIARA 内の AVF
サイクロトロンを利用し、12C6+:320 MeV, 1, 2, 3 Gy
を15シャーレ照射した。

再生した植物体は順次発根、順化、鉢上げを行い、開
花させて花色の確認を行った。色彩値（Hunter b値）は
色彩色差計（CR-321、MINOLTA）で測定した。

その結果、葉片からの再生個体数は、1・2Gyを照
射したものには同程度だったが、3 Gyを照射したものは、
1・2Gyと比較して半分程度であった(Table1)。

花弁から伸長したシュートは、照射線量に関係なく
ビトリフィケーションが多く発生したが、発根培地に
フロリアライトを利用することにより、約1カ月後に
は、症状はほぼ改善され、鉢上げが可能となった。

鉢上げした再生個体の開花は、早くなる傾向を示
ており、1、2Gy照射のものは、1Gy照射したものより
開花が早くなる傾向があった。再生個体数は、1Gy
照射のものは、2Gy照射のものより約2倍であった。

選抜した5個体は、プレートで比較して切り前の黄色
が濃くなり(b値)、黄色スプレーパレットと同等の濃さであ
りが、開花時に外弁が退色するため、黄色品種として
利用することは困難だと思われた(Table2, Fig.1)。今後も、
再生個体の開花処理と調査を繰り返し行い、選抜
濃黄化個体への再照射を検討していきたい。

Reference
1)鹿児島県バイオテクノロジー研究所成績概要書

Table 1. The number of regenerated and selected plants

<table>
<thead>
<tr>
<th>Tissue</th>
<th>No. of plants</th>
<th>12C6+ :Dose (Gy)</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Leaf Regenerated plants</td>
<td>520</td>
<td>492</td>
<td>233</td>
</tr>
<tr>
<td>Selected plants</td>
<td>0</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>Petal Regenerated plants</td>
<td>23</td>
<td>41</td>
<td>73</td>
</tr>
<tr>
<td>Selected plants</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 2. Changes of color indices b

<table>
<thead>
<tr>
<th>Individual</th>
<th>Dose (Gy)</th>
<th>Bud</th>
<th>Full Bloom</th>
</tr>
</thead>
<tbody>
<tr>
<td>Light yellow ‘Jinba’</td>
<td>20.84</td>
<td>23.67</td>
<td>1.57</td>
</tr>
<tr>
<td>No.8</td>
<td>37.25</td>
<td>35.49</td>
<td>1.40</td>
</tr>
<tr>
<td>No.10</td>
<td>37.50</td>
<td>37.30</td>
<td>2.80</td>
</tr>
<tr>
<td>No.11</td>
<td>43.33</td>
<td>34.78</td>
<td>10.27</td>
</tr>
<tr>
<td>No.12</td>
<td>43.07</td>
<td>25.94</td>
<td>16.47</td>
</tr>
<tr>
<td>No.13</td>
<td>37.94</td>
<td>31.07</td>
<td>4.61</td>
</tr>
</tbody>
</table>

Fig.1 Selected flower color mutants
A : Light yellow Jinba  B : No.11  C : No.12  D : No.13
Upper : Bud Lower : Flower
3-16 Ion Beam Breeding of Flower Color Variations in Transgenic Plants with Multi-Disease Tolerance

M. Okamura a), A. Shimizu a), S. Watanabe a), Y. Hase b), I. Narumi b) and A. Tanaka b)

a) Plant Research Center, Kirin Agribio Co., LTD., b) Radiation-applied Biology Division, QuBS, JAEA

1. Introduction

Ion beams have different effect from that of electron beams on mutation generation of crops and have great impact on plant seed and seedling business. New carnation variety series with 10 flower color variations have been developed by ion beam breeding in the joint R&D between Kirin Agribio Co., LTD. and JAEA. They have been commercialized in Japan and Europe

We are trying to develop an advanced application system of ion beams to produce color variations or to add improvements in transgenic plants that have acquired useful characteristics such as tolerance to multi-diseases by recombinant DNA techniques.

2. Materials and Methods

Agrobacterium-mediated transgenic chrysanthemum plants (Dendranthema grandiflora) expressing double-stranded RNA-specific ribonuclease gene (pac1) derived from Schizosaccharomyces pombe were used as materials for ion beam breeding. Leaf segments were placed in petri dishes containing Murashige and Skoog medium supplemented with 0.05 mg/l NAA and 0.1 mg/l BA, 30 g/l sucrose and 7 g/l agar. The samples covered with Kapton film were irradiated with 220 MeV or 320 MeV carbon ion beams from the TIARA AVF cyclotron (JAEA, Takasaki). In vitro plants derived from ion beam irradiation have been acclimatized and cultivated in the greenhouse and their flower color and shape were investigated.

3. Results and Discussions

Transgenic chrysanthemum plants expressing pac1 gene showed tolerance against Chrysanthemum Stunt Viroid (CSVd) and Tomato Spotted Wilt Virus (TSWV). The tolerance of pac1 transgenic chrysanthemum to CSVd is shown in Figure 1.

Thus far 400 plants derived from 3 Gy to 10 Gy irradiation with 220 MeV or 320 MeV carbon ion beams have been cultivated in the greenhouse and flowered. Flower color mutants such as pale pink, dark pink, salmon etc. have been obtained (Figure 2).

The control of viroid disease has been very difficult because factors essential for viroid survival are indispensable to the host plants. Viruses belonging to the genus Tospovirus including TSWV are very serious pathogens not only in chrysanthemum but on other horticultural plants. Ion beam breeding is effective to produce color variations, once the transgenic plants with virus- and viroid-resistance have been acquired.

From these results we confirmed that the advanced application system of ion beams will contribute to the development of variety series of ornamentals with high quality at lower cost, as compared with the method to introduce useful characteristics into several varieties with different flower color by transformation one by one.

References
Mutation Induction on Oriental Hybrid Lily Irradiated with Ion Beams

S. Chinone a), A. Ishizawa a), K. Tokuhiro a), K. Nakatsubo a), M. Amano a), Y. Hase b), I. Narumi b) and A. Tanaka b)

a)Kaneko Seeds CO., LTD., b)Radiation-Applied Biology Division, QuBS, JAEA

To obtain mutants of Oriental hybrid Lily, we investigated the influences of ion beams irradiation to bulb scales in vitro. Survival rates of scales irradiated with 320 MeV carbon and 50 MeV helium ion decreased at 0.2 and 1.5 Gy, respectively. The suitable dose was estimated to be around 0.2–0.4 Gy in carbon ion and 1.5–2.0 Gy in helium ion. Leaf color mutations which chlorophyll were defected partly or wholly were observed.

照射したりん片の生存率や生育量は無照射のりん片に比べて低下し、その影響は\(^{12}\text{C}^+\)照射の方が\(^{4}\text{He}^+\)照射より大きかった。照射したりん片から形成された小球根を817個体、\(^{4}\text{He}^+\)では641個体を定植したところ、クロロフィルが完全にまたは部分的に欠損している個体が観察された（Table.1, Fig.2）。今後は球根養成を行い、花色、花形や雄性不稔等の特性について調査する予定である。

<table>
<thead>
<tr>
<th>Dose (Gy)</th>
<th>No. of bulbs</th>
<th>No. of leaf mutants</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>28</td>
<td>0</td>
</tr>
<tr>
<td>(^{12}\text{C}^+) 0.1</td>
<td>200</td>
<td>0</td>
</tr>
<tr>
<td>(^{4}\text{He}^+) 0.5</td>
<td>126</td>
<td>0</td>
</tr>
<tr>
<td>0.2</td>
<td>14</td>
<td>1</td>
</tr>
<tr>
<td>0.4</td>
<td>20</td>
<td>1</td>
</tr>
<tr>
<td>0.6</td>
<td>14</td>
<td>0</td>
</tr>
<tr>
<td>0.8</td>
<td>59</td>
<td>0</td>
</tr>
</tbody>
</table>

Table.1. The number of leaf color mutants induced by ion beam irradiation.

Fig.1. Influence of 320 MeV carbon ion beam irradiation to bulb scale on survival rate in Lily.

Fig.2. Leaf color mutants induced by ion beam irradiation

Reference
Mutation Breeding on the Ornamental Plants of *Gypsophila* and *Gentiana* Species


*Oita General Service Co., Ltd., **Radiation-Applied Biology Division, QuBS, JAEA

*Gypsophila paniculata* and *Gentiana trifida* var. *japonica* are the important ornamental crops for cut flower use in Japan. In an attempt to induce mutation for breeding use, shoots of such plants on LS medium were irradiated with carbon ion beams using the JAEA AVF cyclotron. Irradiation experiments were conducted two times, and survival rates of the regenerated plants were evaluated when they were transplanted to the pots. The doses suitable for mutation induction are estimated to be 2-4 Gy for *Gypsophila* and 0.5-1 Gy for *Gentiana*. Apparently, *Gentiana* is more susceptible to ion beam irradiation than *Gypsophila*.

Fig. 1 Survival rates of the plants regenerated from carbon ion irradiated shoots of *Gypsophila* and *Gentiana* plants, measured when they were transplanted to pots.

Reference

Breeding New Varieties of Miniature Cymbidium Using Ion Beam Irradiation

S. Yuki a), S. Araki a), T. Suzuki a), T. Ohsuga a), K. Katayama a), Y. Hase b) and Y. Yokota b)

a) Central Research Institute, Ishihara Sangyo Kaisha, Ltd.,
   b) Radiation-applied Biology Division, QuBS, JAEA

The protocorm like bodies (PLBs) of two novel varieties of miniature hybrid Cymbidium, which were bred by crossing Cymbidium hybrida with Cymbidium goeringii, were irradiated with 320 MeV $^{12}$C$^+$ ions at various doses ranging from 0 to 128 Gy. Irradiated PLBs were cultured at 25 °C with a 16 hr photoperiod. After 12 months of culture, the efficiency of regeneration from the PLBs was evaluated. In both varieties, it was drastically decreased at 16 Gy or more. Based on this result, we irradiated PLBs of three varieties of miniature hybrid Cymbidium at doses of 0-12 Gy in a mutation breeding experiment.

3-19 Breeding New Varieties of Miniature Cymbidium Using Ion Beam Irradiation

S. Yuki a), S. Araki a), T. Suzuki a), T. Ohsuga a), K. Katayama a), Y. Hase b) and Y. Yokota b)

a) Central Research Institute, Ishihara Sangyo Kaisha, Ltd.,
   b) Radiation-applied Biology Division, QuBS, JAEA

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はじめに

シンビジウムは、ファレノプシスやデンドロビウムと共に、我国で最も重要なラン科園芸植物の一つとなっている。しかし、近年、業務用花卉需要の減少を受けて、大型鉢物中心のシンビジウム市場は減少傾向にある。この市場の改善を目指し、石原産業㈱では、シンビジウムとシュンランの交配により育種された数種の小形シンビジウムを導入し、生産・販売することを検討している。この小形シンビジウムは、その大きさのため、交配による育種には制約がある。そこで、さらなる品種拡充のため、花卉育種で実績が高く、また、シンビジウムでの変異体作出の報告のある、「イオンビーム育種」の本花卉への適用の検討を始めた。

材料および方法

感受性検定試験1

2種の小形シンビジウム（S-26-9及びS-27-1）より調製したPLB（直径2~3 mm）をメスで2分割し、寒天培地上へ置床した。約1週間、25 ℃、16 hr照明下で培養後、炭素イオン（$^{12}$C$^+$、320 MeV）を照射した。照射線量は、0、0.5、1、2、4、8、16、32、64及び128 Gyとし、一群50個の切片に照射した。照射サンプルは、新しい培地へ移植し、12ヶ月間培養した。その間に、出芽・発根したPLBを再分化したPLBとしてカウントし、0 Gy照射群の再分化率で除した値を各群の再分化率とした。

感受性検定試験2

4種の小形シンビジウム（S-26-1、S-26-9、S-27-1及びS-27-6）のPLBから厚さ1 mm程度の切片を調製し、寒天培地上へ置床した。1~5日間培養後、試験1と同様に炭素イオンを照射し、培養した。

変異誘発試験

3種の小形シンビジウム（S-27-1、S-27-6及びS-39-2）のPLBから厚さ1 mm程度の切片を調製し、寒天培地上で3~4日間培養後、炭素イオンを照射した。照射線量は、0、0.25、0.5、1、2、4、6、8、10及び12 Gyとし、一群100個の切片に照射した。

結果及び考察

感受性検定試験1

2種の小形シンビジウム共に、1 Gy以上の照射で、明確な線量依存的な生育抑制が観察された（データなし）。また、再分化率に関しては、8 Gyまでは僅かな低下のみだったが、16 Gy以上で大きく低下した（Fig.1）。ただし、128 Gyでも、約10%のPLBが再分化しており、本試験においては、照射サンプルの厚さにバラツキがあり、イオンビームが完全に透過していないサンプルが生じたことが想定された。

感受性検定試験2

試験1において、サンプルの厚さに懸念が生じたため、試験2においては、サンプル調製時に、2分割ではなく、削ぎ切りで厚さを1 mm程度に調整した。また、照射前後の培養期間も短縮し、品種も2種追加した。これらの照射サンプルは、現在実験観察中であるが、これまでのところ、試験1とほぼ同様な経過を示している。ただし、やや感受性高くなっていると思われる。

変異誘発試験

感受性検定試験の結果から、変異誘発のための照射線量は16 Gy未満が適当と推定された。そこで、3種の小形シンビジウムのPLBに対して、12 Gy以下の10段階の線量で照射を実施した。今後、これらの照射サンプルから再分化した個体を開花期まで栽培し、優良品種の選抜を試みる予定である。

Fig.1 Effect of ion beams on regeneration rate of PLBs

Reference

1) K. Furukawa, et al., イオンビーム育種研究会第2回大会要旨集.(2005)
The carbon ion beams were widely used as the mutagen for breeding technology and also used for cancer therapy. However, precise effect of ion beams to DNA molecules has not been elucidated.

The molecular mechanism of the mutagenesis caused by carbon ion beams was studied. The mutation sites caused by ion beams were determined and the mutation spectrums were compared with those induced by gamma ray.

*S. cerevisiae* strains used in this study were S288C (RAD+, rad50 and rad52). RAD50p is a component of the nonhomologous end joining repair pathway. RAD52p is a component of the homologous recombination repair pathway.

The yeast cells were irradiated with carbon ions (\(^{12}\text{C}^{5+}\); 220 MeV) with the dose 10 to 300 Gy, and LET was 107 keV/\(\mu\)m. Carbon ion beams were generated from AVF cyclotron in JAEA. The survival rates following irradiation were determined on the basis of colony-forming ability. And, selection of *ura3* cells was accomplished by plating the cells on the media containing 5-fluoroorotic acid\(^1\). The mutation sites of *ura3* mutants were determined by DNA sequencing.

Figure 1 shows the survival rates of several strains exposed to \(^{12}\text{C}^{5+}\) ion. *rad50* and *rad52* cells showed high sensitivity to ion-beam (Fig.1).

The *URA3* regions (804 bp) were sequenced. Fig. 2 shows the mutational spectrums of irradiation ion beam and gamma ray. The remarkable feature of mutations in wild type cells induced by carbon ions was that the mutations sites were localized near the linker regions of nucleosomes\(^2\). On the contrary, the mutations were induced in the DNA region which combined with the histone-protein in *rad50* or *rad52* strain (Fig.2). This difference may result from the repair pathway which operates in mutant strains. Moreover, the GC \(\rightarrow\) TA transversion was largely observed. It suggests that the mutations by ion beams resulted from oxidative damage such as formation of 8-oxodGTP mainly\(^{3}\). To elucidate the ion beam effect on the budding yeast, we are going to analysis *ogg1* and *msh2* mutant strains which are deficient in oxidized purine base lesion DNA N-glycosylase activity and mismatch repair activity, respectively.

References
3) P. M. Girard et al., Carcinogenesis. 19 (1998) pp1299-1305

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**Fig. 1.** Survival curves for yeast strain S288c after irradiation with carbon ion beams.

**Fig. 2.** Mutational spectrum of irradiation ion beam (above) and gamma ray (below).
3-21 Mutation Induction in Azalea Using Ion Beam Irradiation and Its Gene Analysis

N. Kobayashi a), S. Sakamoto a), A. Nakatsuka a), Y. Hase b) and I. Narumi b)

a)Faculty of Life and Environmental Science, Shimane University
b)Radiation-Applied Biology Division, QuBS, JAEA

1. Introduction
In this study, ion beams were irradiated to azalea seeds and cultured leaf segments to induce flower color and shape mutation. Additionally, we try to research the key genes of ion beam induced mutations as a final purpose; anthocyanin biosynthesis genes related to flower color mutation and homeotic genes related to flower shape and organ mutation that we already isolated 1,2).

2. Materials and Methods
2-1. Seeds of *Rhododendron kaempferi*, *R. ripense*, *R. japonicum* and *R. matternichii* var. *brevitolium* were irradiated with carbon ion beam (220 MeV $^{12}$C$^{5+}$) at 0 to 50 Gy (Fig. 1). After the irradiation, seeds were sowed on sphagnum moss bed in pot. The germination rate was evaluated after 6 weeks.

2-2. Young leaves of potted *R. × pulchrum* ‘Sen-e-oomurasaki’ were excised, sterilized and used as explants materials as previous study3). The leaf segments were cultured on modified 1/2 Murashige and Skoog’s medium supplemented with 2 μM NAA and 9 μM TDZ in the test tube4). The explants were transplanted on the high concentration agar medium in the tissue culture dishes and covered with Kapton film. The transported samples were irradiated with carbon ion beam (220 MeV $^{12}$C$^{5+}$) at various doses (0 to 128 Gy, Fig. 2). The leaf segments were transplanted on the initial medium and cultured at 22±1 °C with 16 hours-photoperiod after the irradiation. The regeneration rates of callus, shoot primordia and shoot were investigated after 2 months.

3. Results and Discussion
3-1. In preliminary experiment, seeds of 5 species were irradiated with carbon ion beam at ranges from 0 to 100 Gy5). These seeds were difficult to germinate with ranges more than 60 Gy irradiation. Therefore, the second irradiation was applied narrow doses (0 to 50 Gy) in the same condition (Fig. 1). The germination rates were strongly downed at the ranges from 30 to 50 Gy and different depending species. The optimal irradiation doses for seeds considering from median germination decrease may be 30 to 40 Gy for *R. kaempferi*, 20 Gy for *R. ripense*, 10 to 20 Gy for *R. japonicum* and 20 to 30 Gy for *R. matternichii* var. *brevitolium*.

3-2. The regeneration rate of callus decreased according to the ion beam strength from 8 Gy irradiated leaves (Fig. 2). While, shoot primordia and shoot were not able to regenerate with more than 8 to 16 Gy irradiation (Fig. 2, 3). The effective conditions to obtain irradiated mutants were thought to be from 4 to 8 Gy exclusive for *R. × pulchrum* ‘Sen-e-oomurasaki’ leaves. Mutation research will determine plants regenerated from these shoot primordia and shoot of ion beam irradiation.

4. References

Fig. 1 Dose response of germination rate in 4 species at 6 weeks after sowing.

Fig. 2 Regeneration rates of callus, shoot primordia and shoot from *R. × pulchrum* ‘Sen-e-oomurasaki’ leaf segments at 2 months after irradiation.

Fig. 3 Dose response of regeneration from *R. × pulchrum* ‘Sen-e-oomurasaki’ leaf segments at 2 months after irradiation.
3-22 Effects of Ion Beam Irradiation on Plant Growth and Morphology in Soybean

A. Kanazawa a), J. Abe a), Y. Hase b) and A. Tanaka b)

a) Research Faculty of Agriculture, Hokkaido University, b) QuBS, JAEA

Soybean (Glycine max) is an important crop in terms of production of food, oil, and forage. However, existing mutant lines of soybean are very limited, which is a constraint on performing genetic study and breeding of this plant. One of the reasons for the small number of mutant lines in soybean appears to be associated with difficulty in production of a mutant and to duplication of genomic segments. This plant is considered to have derived from ancestral plant(s) that have a tetraploid genome, and as a consequence, more than 90% of nucleotide sequence in the soybean genome is duplicated. It is conceivable that such a duplicated nature of the genome brought about a low frequency of mutant production by conventional methods for mutagenesis such as γ-ray or X-ray irradiation as well as chemical treatments. In these circumstances, we have started to examine whether ion beam irradiation is effective in producing a mutant in soybean because ion beam irradiation is expected to cause genomic changes that are more drastic than those induced by conventional mutagenesis.

Dried seeds of soybean were exposed to the 320 MeV carbon ions with a range of 0.2-25 Gy. In order to irradiate at an equal dose on seeds, seeds were fixed on a plastic dish in which meristem tissues were positioned on the top (Fig. 1). The irradiated seeds were sown on soil and plants were grown for three weeks in a greenhouse. A prominent decrease in plant height was observed depending on the dose in the range of 5-25 Gy (Fig. 2), but not in the range lower than 5 Gy (0.2, 0.5, 1.0, and 2.0 Gy; data not shown), indicating that doses higher than approximately 5 Gy affect plant growth rate.

In addition to the inhibition of plant growth, morphological changes were also induced by irradiation. One of the prominent changes in morphology was detected in the primary leaves in which a narrow portion was generated in the central part of a leaf. This type of change was detected in 15 plants per 66 plants irradiated at 5 Gy, and in 4 plants per 56 plants irradiated at 2 Gy. The other prominent morphological change was unscheduled generation of stem(s) from the node where cotyledons were formed (Fig. 3). This type of change was detected in 8 plants per 66 plants irradiated at 5 Gy, and in 2 plants per 56 plants irradiated at 2 Gy. Both types of change were not detected in plants irradiated at the doses lower than 1 Gy.

Based on the observed effects of irradiation at various doses on plants, we concluded that irradiation at 2-5 Gy is most appropriate for producing mutant population in our material. We subsequently performed a large-scale irradiation to soybean seeds at the doses of 5 Gy and 2.5 Gy to 3,200 seeds and 3,320 seeds, respectively, and grew them in the field of Hokkaido University. We are currently analyzing the effects of irradiation using field-grown plants.

References

Fig. 1 Soybean seeds placed on a dish for irradiation.

Fig. 2 Growth of plants after irradiation at the doses of 5-40 Gy. Average ± SE are shown.

Fig. 3 An altered phenotype of plants as a consequence of irradiation. Left, control plant; right, plant exhibiting altered morphology.
Mutation Induction by Carbon Ion Beam Irradiation in Banana (Musa spp.)

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Life and Environmental Sciences, University of Tsukuba,
Estación Experimental Tropical Pichilingue, Ecuador,
Radiation-Applied Biology Division, QuBS, JAEA

In Banana (Musa acuminata AAA), most research focuses on generating cultivars tolerant/resistant to black Sigatoka disease caused by Mycosphaerella fijiensis Morelet which is the most dangerous disease distributed worldwide. Ion beams radiation breeding permitted us to select some candidates plants at the nursery condition showing some tolerance against to this disease; however, others interesting characteristics were also observed.

The irradiation was conducted at the Takasaki Ion Accelerators for Advanced Radiation Application (TIARA), Japan Atomic Energy Agency (JAEA). Carbon ion beams irradiation was applied on in vitro explants of banana cultivars ‘Cavendish Enano’ and ‘Williams’ with the dose of 0, 0.5, 1, 2, 4, 8, 16, 32, 64 and 128 Gy. Recorded data were: survival rate (%), relative DNA content (by flow cytometry), leaf disc necrotic area (LDNA-%) under the juglone toxin inoculation, disease development period (DDP-days) and infection index (II-%) by using the fungus of Mycosphaerella fijiensis Morelet for inoculation.

Regarding to the survival rate (%) in general terms, the higher irradiation dose resulted in higher mortality rate, but doses of 8 Gy and lower did not seem to have any effect on survival rates. The flow cytometer analysis detected hexaploid cells among the samples (Fig. 1A). ‘Cavendish Enano’, and ‘Williams’ reported 5 and 1 hexaploid plants, respectively at 4 Gy. This could be a way of causing chromosome duplication, efficiently and fast, especially on improved diploids possessing resistance that could be used as parental lines for banana breeding. A fast growing plantlet (Fig. 1B) was also observed among the irradiated cultivar which is a very important characteristic as it may result in earliness of fruit harvesting.

For selecting tolerant/resistant plants to black Sigatoka, the variables DDP-days, II-% and LDNA-% were combined by linear regression to categorise the response to black Sigatoka in the irradiated materials. LDNA-% regression versus II-% showed high significance (p ≤ 0.01) compared with LDNA-% versus DDP-days, and DDP-days versus II-% that were significant at the 5% level (p ≤ 0.05) in ‘Williams’ (Fig. 2). Six plants in ‘Williams’ and two plants in ‘Cavendish Enano’ were selected as candidates with increased tolerance to black Sigatoka disease. Regarding the complete assessment of the candidate plants, field experiments based on the whole plant cycle are necessary to evaluate not only the response to black Sigatoka disease but also fruit quality, potential production and post-harvest parameters as valuable components for final selections.

Fig. 1. A. Hexaploid cells (encircled) in ‘C. Enano’ produced by ion beam dose 4 Gy. B. ‘C. Enano’ at 4 Gy showing a fast growth (in the center).

Fig. 2. LDNA-% regression versus II-% (A), LDNA-% regression versus DDP-days (B) and DDP-days regression versus II-% (C) in ‘Williams’. Encircled bold and numbering triangles belong to the selected candidate plants that clearly show relationships.
ラベンダーのエッセンシャルオイルは多くの化合物で構成されており、その成分比の違いにより香調が大きく変化することから新しい香りの創出が期待されている。

本研究では、\(^{12}\)C\(_{6}^{+}\)ビームの照射により、エッセンシャルオイル成分比が親植物体と大きく異なり香調が変化した変異体の形成を目的として、\(^{12}\)C\(_{6}^{+}\)ビームの線量がラベンダー培養細胞の増殖および不定芽の形成に及ぼす影響を調査した。

植物材料としてラバンジン（真正ラベンダーとスパイクラベンダーとの雑種）品種「スーパーセビリアンブルー」を用いた。0.2 mg/l 2,4-Dと0.2 mg/lカイネチンを添加したMS固形培地で誘導した葉由来カルスを同組成の液体培地で振盪培養し、得られた懸濁培養細胞を細胞密度が2×10\(^{5}\)個/mlとなるように調整した。カルス培養と同組成の固形培地に入れた直径6 cmのプラスチックシャーレに細胞懸濁液を0.5 ml分注し、320 MeV \(^{12}\)C\(_{6}^{+}\)ビームを0, 0.5, 1.0, 2.0, 4.0, 8.0, 16.0, 32.0, 64.0および128.0 Gyの線量で照射した。照射後、25℃、暗条件で培養を行い、4週間後に増殖したカルスの大きさを0から5の6段階で評価した。評価後、カルスを1mg/l BAを添加したMS固形培地に移植して25℃、明条件で培養し不定芽の誘導を試みた。実験は4回反復で行った。

Fig.1 カルスの増殖程度を評価したところ、0 Gy区ではカルス形成が見られない培地は認められなかったものの、0.5 Gy区および1.0 Gy区も0 Gy区とほぼ同様の指数を示していたが、2.0 Gy以上では全くカルスの形成が認められない。
Screening of Higher Astaxanthin Producing Mutants of a Green Unicellular Alga *Haematococcus pluvialis* by Ion Beam Irradiation

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A highly anti-oxidative ketocarotenoid, astaxathin has been produced by culturing a unique green unicellular alga *H. pluvialis* in large-scale photobioreactors at rich solar energy areas such as Hawaii islands. Since it seems disadvantageous to carry out the algal culture phototrophically in a commercial basis in Japan, we have applied heterotrophic cultivation for the alga. In the dark condition, the alga was found to have a very limited carbon source specificity, namely acetate, and produce astaxanthin about one-third of the phototrophic condition (20 mg-astaxanthin/l) as the cellular content basis (2% w/w). In this study, we have examined the possibility of ion beam irradiation technology for the alga to obtain higher astaxanthin producing mutants, and also high tolerant mutants to acetate. Firstly, the dose effect of ion beam was studied for two culture conditions. In addition, several algal cell preparation techniques including filtration on paper, glass wool, or nitrocellulose, and spreading directly on agar were tested for the better cell recovery.

References


3-26 Role of LexA2 in Radiation Response Mechanism of Deinococcus radiodurans

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Introduction
RecA and LexA proteins in Escherichia coli play important roles in the inducible DNA damage response and repair mechanism (the SOS system). Following DNA damage, RecA is activated by binding to single-stranded DNA which is generated by DNA damage. The interaction of activated RecA and LexA results in the proteolytic cleavage of LexA. RecA-mediated cleavage of LexA inactivates LexA as a repressor. Consequently, various DNA repair genes controlled under LexA are derepressed, thereby the intracellular levels of various DNA repair proteins including RecA are transiently increased.

Although the radioresistance bacterium Deinococcus radiodurans possesses LexA1 protein which resembles E. coli LexA protein, LexA1 is not involved in induction of DNA repair proteins following DNA damage1). Bioinformatic analysis of the D. radiodurans genome sequence showed that the genome encodes a second, diverged copy of LexA1, named LexA2. In an effort to gain an insight into the role of LexA2 in the radiation response mechanism, recA, lexA1 and lexA2 disruptant strains were generated, and the sensitivity to γ-rays, the expression of radiation inducible DNA repair gene and the changes in intracellular level of protein following irradiation were investigated.

Experimental procedures
Gene disruptants were generated using directed insertional mutagenesis technique that was developed by our group2, 3). D. radiodurans cells were resuspended in 10 mM sodium phosphate buffer. The cell suspension was irradiated with 60Co γ-rays at dose rates from 0.1 to 5 kGy/h at Food Irradiation Facility, IAEA. The gene expression and the changes in intracellular level of protein were analyzed by dual luciferase assay4) and western blotting method5), respectively.

Results and discussion
Like LexA1, LexA2 was cleaved by activated RecA to produce two breakdown products. The intracellular level of LexA2 in the recA disruptant strain did not change following γ irradiation, indicating that RecA is the sole protein required for LexA2 cleavage.

RecA induction following γ irradiation was also observed in the lexA2 disruptant and lexA1 lexA2 double-disruptant strains, clearly indicating that neither LexA1 nor LexA2 is involved in RecA induction. Interestingly, as shown in Fig. 1, the lexA2 disruptant and lexA1 lexA2 double-disruptant strains exhibited a much higher resistance to γ-rays than the wild type and lexA1 disruptant strains. Since the decrease in the intracellular LexA1 level reflects the co-protease activity of activated RecA, the recovery to a basal level of LexA1 indicates the inactivation of RecA. The inactivation of RecA was faster in the lexA2 disruptant strain than in the wild type strain. Given that the inactivation of RecA is associated with the completion of DNA repair, the DNA repair capacity might be enhanced in the lexA2 disruptant strain relative to the wild type strain.

Furthermore, pprA promoter activation was enhanced in the lexA2 disruptant strain following γ irradiation. The pprA gene encoding the novel radiation-inducible protein PprA plays a critical role in the radioresistance of D. radiodurans6). The increase in radioresistance of the lexA2 disruptant strain is explained in part by the enhancement of PprA functions7).

References
4) H. Ohba et al., Gene 363 (2005) 133.

![Fig. 1 Survival curves for γ rays. Symbols: closed circles, wild type; open squares, lexA1 disruptant strain; open triangles, lexA2 disruptant strain; closed lozenges, lexA1 lexA2 double-disruptant strain.](image)
3-27 Analysis of Mutagenic Effect Induced by Ion Beams for Breeding of Aspergillus oryzae

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In this study, we investigated the mutagenic effect induced by ion beams for breeding of A. oryzae, which is used in the manufacture of traditional Japanese fermented foods, such as soy sauce, sake and miso. We irradiated A. oryzae with 220 MeV carbon ions by AVF cyclotron at the doses of 300-700 Gy. Mutation rate was measured by counting the number of selenic acid resistant colonies whose mutation relates to sB and sC genes that are involved in sulfur incorporation. These analyses indicated the suitable dose for obtaining variants to be 400 Gy. Furthermore, PCR analysis suggested 57% of these variants carry a deletion with sC or sB gene under this condition.

【緒言】
麹菌（Aspergillus oryzae）は醤油、清酒、味噌などの醸造産業において利用される重要な微生物である。そのため古来より麹菌の育種が精力的に行われてきた。近年、植物などの育種においてイオンビームが注目されており、その変異効果が従来の変異原に比べて高く、変異スペクトラルも様々であることが報告されている。そこで我々は、麹菌育種の新しい手段としてイオンビームの利用を検討することとした。麹菌へのイオンビーム照射についてのデータはまだ少数であることから、まず、照射条件や変異効果について検討した。

【実験方法】
・サンプル調製および照射条件
  A. oryzae (niaD300)の分生子を凍結乾燥し、照射サンプルとした。照射はTIARAのAVFサイクロトロンを用いて加速した^{12}C^{5+} (220 MeV, 107 keV/μm)により行った。
  ・生存率と変異率
    生存率はイオンビームを300～700 Gy照射したサンプルを栄養培地に播種し、生育したコロニー数から生存率を求めた。
    イオンビーム照射サンプルをメチオニン 30 ppm、セレン酸 0.1 mMを含む最少培地に播種し、得られたセレン酸耐性株の数から変異率を求めた。
    ・変異箇所の探索
      硫黄の取り込みに関与するsB遺伝子、sC遺伝子が変異することによりセレン酸耐性になることが知られている1)。sB、sCの前半、後半領域の各断片を増幅するプライマーを設計し、遺伝子欠損の有無についてPCRにより検討した。

【結果】
生存率については線量が大きくなるに従って低下した。また、繰返しごとの変異率は 400 Gyで最大を示した（Fig. 1）。次に、得られた変異株の変異パターンを解析した。変異株のゲノムを抽出しPCRを行ったところ、遺伝子の前半領域のみ、もしくは後半のみが増幅されるもの、両方とも増幅されないものなど、欠損変異を示唆するデータが得られた。これらの株の割合を求めたところ、全耐性株の 27%ほどを占め、高い確率で遺伝子欠損株が得られることが示唆された。また、400 Gy照射区では欠損変異を示唆する株の割合が57%であり、他の照射区に比べ極めて高い割合であった（Table 1）。

以上のことから、凍結乾燥処理を行った麹菌のイオンビーム照射では 400 Gy程度の線量で高い変異効果が期待でき、変異株の欠損変異が高い割合で生じていることが示唆された。

【文献】

![Fig. 1 Survival rate and Mutation rate of conidia after irradiation with ion beams in A. oryzae.](image)

Table 1 Difference in mutagenic effect at various doses of ion beams in A. oryzae.

<table>
<thead>
<tr>
<th>Dose (Gy)</th>
<th>0</th>
<th>300</th>
<th>400</th>
<th>500</th>
<th>600</th>
<th>700</th>
</tr>
</thead>
<tbody>
<tr>
<td>Survival rate (%)</td>
<td>100</td>
<td>4.13</td>
<td>0.31</td>
<td>0.08</td>
<td>0.03</td>
<td>0.01</td>
</tr>
<tr>
<td>Mutation rate (%)</td>
<td>0.028</td>
<td>0.070</td>
<td>0.055</td>
<td>0.018</td>
<td>0.009</td>
<td></td>
</tr>
<tr>
<td>No. of variants (cfu)</td>
<td>0</td>
<td>12</td>
<td>21</td>
<td>11</td>
<td>5</td>
<td>2</td>
</tr>
<tr>
<td>No. of deficient strain (cfu)</td>
<td>0</td>
<td>1</td>
<td>12</td>
<td>1</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Percentage of deficient strain per variants (%)</td>
<td>8.3</td>
<td>57.1</td>
<td>9.1</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Reference
3-28 Mutation Breeding of Koji Mold Induced by Ion Beam

K. Ito a), J. Takeichi a), Y. Hanya a), K. Satoh b), Y. Hase b) and I. Narumi b)
a) R&D Division, Kikkoman Corp., b) Radiation-Applied Biology Division, QuBS, JAEA

Introduction

Koji molds, Aspergillus sojae and Aspergillus oryzae, are industrially important microorganisms which is used for brewing of soy sauce. The various mutants have been used for mutation breeding of koji molds for many years. However, there are problems in usual mutants, for example UV, NTG and X-rays lead to much inconvenient mutation such as slow growth rate and poor conidiation. Consequently, most mutants simultaneously have these traits, even if target trait was acquired. Therefore, we focused on mutation by ion beam. It is known that this method induces targeted mutations with smaller number of inconvenient mutation. We previously reported that survival rate and mutation frequency of A. sojae IFO4241 following irradiation of ion beams and determined optimal irradiation conditions. In this report, we investigated whether ion beams induce deletion mutations in A. sojae IFO4241 as well as higher plants. A defect in the wA gene of Aspergillus species is associated with the loss of conidial pigmentation and the conidial color of this mutant will turn to white from green. To investigate the deletion mutation frequency, the pigmentless-conidium mutants were obtained and the wA gene of these mutants were analyzed.

Materials and Methods

(1) Isolation of pigmentless conidium-mutants
The suspension containing 10⁶ conidia (A. sojae IFO4241) were spread on Malts agar plate and incubated at 30 ºC for 6 hr for germination. Germinated conidia were irradiated with 220 MeV ¹²⁷I ions ion beam accelerated by AVF cyclotron at TIARA with the dose of 100, 150 and 200 Gy. Irradiated conidia were cultured at 30 ºC for 3 days. The pigmentless-conidium mutants were picked up and purified three times by single conidium isolation on Malts agar plate containing 0.25 % Triton X-100. UV irradiation was performed as previously described. Only one mutant was picked up from each irradiated plate.

(2) DNA analysis of wA gene of pigmentless-conidium mutants
The full length of genomic wA gene from pigmentless conidium mutants and/or their internal regions were amplified by PCR using various primer sets. The amplified DNA fragment size was compared with that of wild type on 0.8 % agarose gel. Southern blot analysis was performed using DIG labeling Kit (Roche). The PCR fragment of wA gene was used as a probe for southern analysis.

Results and Discussion

The number of pigmentless-conidium mutants from irradiated plates with ion beam and/or UV was 114 and 38, respectively (Table 1). The full-length wA gene (8.0 kb) of pigmentless-conidium mutants was amplified by primer F1 and R2 (Fig. 1). We evaluated the deletion mutation of wA gene if the PCR fragment was not amplified (Fig. 1 (B)-(D)) or the amplified PCR fragment size was smaller than that of wild type (Fig. 1 (A)). Out of 114 pigmentless-conidium mutants with ion beam irradiation, 38 mutants were deletion mutant (Table 1). On the other hand, all pigmentless-conidium mutants with UV irradiation were not due to deletion mutation. These results indicated that deletion mutation frequency with ion beam irradiation was higher than that with UV irradiation. PCR analysis of 38 deletion mutants revealed that the deletion mutation was classified into four patterns (Table 2 (A)-(D)). The loss of whole wA gene of pattern C mutants was confirmed by southern blot analysis (data not shown). This result revealed that an 8.0-kb chromosomal region could be deleted with ion beam irradiation in A. sojae IFO4241. The large deletion mutation might be occurred in A. sojae IFO4241 as well as higher plants.

Table 1 Comparison of deletion mutation frequency between ion beam irradiation and UV irradiation.

<table>
<thead>
<tr>
<th>Beam Type</th>
<th>Number of irradiated plates</th>
<th>Number of pigmentless mutants</th>
<th>Number of deletion mutants</th>
<th>Deletion mutants per pigmentless-conidium mutants (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ion beam</td>
<td>144</td>
<td>114</td>
<td>38</td>
<td>33</td>
</tr>
<tr>
<td>UV</td>
<td>120</td>
<td>38</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Fig. 1 PCR analysis of the wA gene of deletion mutants. The pyrG gene was amplified as a positive control. The lane Mr is DNA size marker.

Table 2 The mutation pattern of wA gene in deletion mutants.

<table>
<thead>
<tr>
<th>Mutation pattern</th>
<th>Number of mutants</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A) Deletion of internal region</td>
<td>22</td>
</tr>
<tr>
<td>(B) Deletion of 3’ region</td>
<td>7</td>
</tr>
<tr>
<td>(C) Deletion of full genomic region</td>
<td>2</td>
</tr>
<tr>
<td>(D) Translocation, inversion etc.</td>
<td>7</td>
</tr>
</tbody>
</table>

References

2) M. E. Mayorga et al., Genetics 126 (1990) 73.
3-29 Deletion of Minor Enzyme Activities of *Rhizomucor miehei* by Heavy Ion Beam Irradiation

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a) Tokyo Research Laboratory, Meito Sangyo Co., Ltd., b) Radiation-Applied Biology Division, QuBS, JAEA

1. Introduction

Cheese is one of the major dairy products and various kinds of cheese are produced in the world. Rennet, which breaks kappa-casein and induces coagulation of milk, is originated from veal of lactating calf or lamb stomach or produced by fermentation of *Rhizomucor (Rh.) miehei*. *Rh. miehei* also produces various minor enzymes such as lipase and these enzymes usually cause production of undesirable flavors during maturation of cheese. For this reason, acidification and chemical treatments are employed to improve the property. However, these treatments also cause loss of coagulation activity. Therefore, breeding a strain of lipase-less or very low lipase activity is expected for improving the productivity and quality of cheese.

The survival rate of spore was about 1% following 200 Gy irradiations of $^{125}$I at 220 MeV and $^{26}$Ne at 350 MeV. On the other hand, He and Ar ions had lower effect on survival rate than those of C and Ne ions. In 2006, lipase-less mutant separation was performed by irradiation of C and Ne ions at doses of 100 and 200 Gy to spore.

2. Materials and Methods

Sporangiospore of two parent mutants of *Rh. miehei* obtained by chemical and $\gamma$-ray mutagenesis was harvested from plate cultivation and was spread on a petridish as previously reported. Spore was dried over silica gel, conditioned over saturated NaCl solution, and covered with polyimide film for the beam irradiation. The beam irradiation of C-220 MeV and Ne-350 MeV was performed at AVF cyclotron of TIARA.

Irradiated spore was resuspended in sterile water, spread over glucose-yeast extract agar plate, and cultivated at 37°C for three days. A mutation rate was determined by a frequency observation of red-pigmented colony that was caused by the mutation in AMP-synthesis pathway and accumulation of polyribosyl-aminomidazole during cultivation at 37°C.1)

After mutation rate determination, the colonies were transfer to agar plate containing milk to detect secreted rennet activity from colonies. Coagulation activity was detected by formation of clear zone on the milk agar plate. After contacting with the milk agar plate overnight, the colonies were transferred onto agar plate containing olive oil to detect lipase activity secretion. Colonies those formed inarticulate clear zone on each medium were cultured to confirm the coagulation and lipase activity production by a tube cultivation with a medium that was consisted of soybean flower and soluble starch. Lipase activity was determined by an alkali-titration released fatty acid from olive oil. Coagulation activity was determined by comparing clotting time (time for initiating coagulation of milk) with a standard enzyme using skimmed milk solution as a substrate.

3. Results and Discussion

Mutation rate varied from 0.06 to 0.99%. A mutagenesis effect of neon ions was lower than that of carbon ions as shown in Table 1.

After observation of the mutation rate, colonies were transferred onto milk-agar and then oil-agar medium to select a deficient mutant by an inarticulate clear zone formation. All mutants isolated on agar media excreted lipase in tube cultivation. Among them, some mutants showed several times smaller activities than that of the parent strain (Table 2). These mutants might be heterozygous and further purification is necessary.

A mutant strain with low coagulation activity was also found and its culture product showed a different chromatographic profile from that of parent (data not shown).

Table 1 Mutation rate by irradiation of heavy ion beams. Irradiated spore was spread on agar plate medium and cultivated for 3 days at 37°C. Mutation rate is defined as the ratio of red-pigmented colonies in total colony.

<table>
<thead>
<tr>
<th>Parent</th>
<th>Irradiation</th>
<th>Total colony</th>
<th>Mutation rate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>B</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>C-100 Gy</td>
<td>252</td>
<td>0.40%</td>
</tr>
<tr>
<td></td>
<td>C-200 Gy</td>
<td>1,313</td>
<td>0.99%</td>
</tr>
<tr>
<td></td>
<td>Ne-100 Gy</td>
<td>1,805</td>
<td>0.06%</td>
</tr>
<tr>
<td></td>
<td>D</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>C-200 Gy</td>
<td>406</td>
<td>0.49%</td>
</tr>
</tbody>
</table>

Table 2 Mutants with low lipase activity.

Mutant strains were isolated from the parent strain B and D irradiated with neon and carbon ions at a dose of 200 Gy. Enzyme activity was shown as a proportional value to the activity of each parent.

<table>
<thead>
<tr>
<th>Mutant</th>
<th>Coagulation activity</th>
<th>Lipase activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>B-14 (Ne-200 Gy)</td>
<td>95%</td>
<td>23%</td>
</tr>
<tr>
<td>B-43 (Ne-200 Gy)</td>
<td>102%</td>
<td>21%</td>
</tr>
<tr>
<td>D-57 (C-200 Gy)</td>
<td>35%</td>
<td>75%</td>
</tr>
</tbody>
</table>

Reference

3-30 Spectral Analysis of DNA Damages Induced by He$^{2+}$ Ion Beam Compared to Those of $^{60}$Co $\gamma$-rays

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1. Introduction

Information in relation to chemical structures and their distribution on DNA is quite important to clarify following biological repair procedures. In case of ionizing radiations, especially, it is expected that three-dimensional distribution of DNA damages is diversified as well as their chemical structures, and the diversity makes investigations of physicochemical processes in radiation effects difficult. In fact, double-strand breaks produced by high linear energy transfer (LET) alpha particles are more difficult to rejoin in living cells than those produced by low LET $^{137}$Cs $\gamma$-rays. To investigate difference of DNA damage spectrum between radiations, we have developed new methodology to analyze strand break-termini and nucleobase lesions using snake venom phosphodiesterase (SVPD), calf intestinal alkaline phosphatase (CIAP) and piperidine. In this report, the results of the DNA damage spectra induced by He$^{2+}$ ion beams and $^{60}$Co $\gamma$-rays are demonstrated. The He$^{2+}$ ion beams used was obtained from TC-1 port of 3 MV tandem accelerator in TIARA at Takasaki Advanced Radiation Research Institute. The $^{60}$Co $\gamma$-rays, as a standard radiation source, were irradiated in the second cell in the first building of $\gamma$-irradiation facility at the same institute.

2. Materials and Methods

Linear formed pUC19 plasmid DNA digested with SmaI, (pUC19/SmaI) was used. pUC19/Smal aqueous solution was mounted on a silicon plate and dried thoroughly in vacuum under $P_2O_5$. For $\gamma$-irradiation, the dried DNA sample on a plate was put into a glass tube sealed in vacuum to achieve secondary electron equilibrium. Each sample was irradiated at a dose (dose rate) of 40 (0.333), 100 (0.833), 400 (3.33), 900 (7.50), and 1,800 kGy (15.0 kGy/h) at r.t., respectively. For He$^{2+}$ beams, the DNA-mounted plate was fixed in a cylindrical chamber (39 mm$^3$, hight: 22 mm) with a Kapton® window (8 $\mu$m) under argon gas. Each sample was irradiated with ~200 keV/µm of the beam just under the Bragg-maximum, which was controlled using a depth-tunable cell irradiation equipment, at a dose (dose rate) of 100 (1.068), 400 (1.068) and 900 kGy (1.068 kGy/pass) at r.t.. The irradiated DNA samples were recovered by water at 0°C to be 0.32 $\mu$g/µL.

The new method for DNA damage analysis is introduced in previous paper in detail. In brief, the solution dissolving irradiated DNA was treated directly by SVPD, or was treated by SVPD following to CIAP pretreatment, to produce 5'-deoxynucleotides (pdNs) from 3' termini without phosphate for 15 min at 31.4 °C. The pdNs were quantified by HPLC after dephosphorylation to be 2'-deoxynucleosides by additional CIAP treatment. Furthermore, the irradiated DNA sample was pretreated by piperidine, to convert a variety of nucleobase damages and abasic sites into 3' termini with phosphate. The increase of 3' termini with phosphate by piperidine treatment reflects the amount of piperidine-labile nucleobase lesions and abasic sites. Based on the series of the experiments, the yields of total 3' termini, those of 3' termini with or without phosphate, and piperidine-labile lesions were calculated. (Table 1).

3. Results and Discussion

Table 1 shows the yields of DNA lesions calculated. Additionally, the yields of unaltered nucleobase release are also demonstrated as a reference. In case of the strand break patterns, the yields of them for He$^{2+}$ were almost the same as those for $\gamma$-rays. In general, 3' termini with phosphate were predominantly produced compared with those without phosphate. On the other hand, in case of piperidine-labile lesions, the yields of them for He$^{2+}$ were twice as large as those for $\gamma$-rays. This tendency was shown also in case of unaltered nucleobase release. Although the yields of nucleobase lesions for high-LET radiations would be larger than those for low-LET radiations according to the data from the table, it is also possible that piperidine-labile sites are susceptible to be produced in the high-LET radiation field. Anyway, more investigation additionally using base-excision enzymes should be needed to clarify the difference between the radiations. Moreover, the differences in DNA spectrum between on and off peaks of the Bragg-maximum, and between some degrees of hydration of DNA molecule are to be investigated.

<table>
<thead>
<tr>
<th>Category of lesions</th>
<th>Yield (µmol U x 10$^3$ Gy/Br2+)</th>
<th>$^{60}$Co (µmol U x 10$^3$ Gy/Br2+)</th>
<th>He$^{2+}$ (~ 200 keV/µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3' termini without phosphate</td>
<td>0.004</td>
<td>0.024</td>
<td></td>
</tr>
<tr>
<td>3' termini with phosphate</td>
<td>0.016</td>
<td>0.058</td>
<td></td>
</tr>
<tr>
<td>Piperidine-labile lesions</td>
<td>0.004</td>
<td>0.183</td>
<td></td>
</tr>
<tr>
<td>Unaltered nucleobase release (ref.)</td>
<td>0.013</td>
<td>0.434</td>
<td></td>
</tr>
</tbody>
</table>

4. Acknowledgments

We would like to gratefully thank Dr. Y. Hase at Gene Resource Research Group in Takasaki Advanced Research Institute for maintaining and operating the TC-1 beamline.

References

Ionizing radiation causes various types of DNA lesions, such as strand breaks, base lesions and, in particular, biologically relevant complex damage known as cluster damage sites, which consist of two or more elemental lesions within one or two helical turns of DNA. Clustered DNA damage sites are considered to be less readily repaired than isolated lesions and therefore may induce deleterious and serious genetic changes in cells. The complexity of clustered damage is proposed to increase on increasing the ionizing density of the radiation (LET). It has been demonstrated that the direct effect of low LET radiation produces 30-40% of the lesions in cells. This value is shown to reach 70% when cells are irradiated with α-particles. However, few investigations have been systematically undertaken to elucidate the nature of lesions produced by the direct effects of radiation with varying ionizing density.

To gain insights on the nature of lesions induced by the direct effect of the radiation, yields of single- and double-strand breaks (ssb’s and dsb’s), and base lesions for hydrated plasmid DNA were measured after exposure to carbon ion particles with varying LETs. The use of hydrated hydrated plasmids becomes more readily clustered on increasing the oxidative base lesions into an ssb.

With two glycosylases, Nth and Fpg proteins, which convert the oxidative base lesions to ssb, the yield of prompt ssb after exposing hydrated plasmids to carbon ion particles with a LET of 87 keV/µm was estimated to reach 70% when cells are irradiated with carbon ion particles with varying LETs. The use of hydrated DNA eliminates the indirect effect of radiation arising from irradiation of bulk water. Base lesions were detected as additional strand breaks after treatment of irradiated DNA with two glycosylases, Nth and Fpg proteins, which convert the oxidative base lesions into an ssb.

The yield of prompt ssb after exposing hydrated plasmid DNA to carbon ion particles with a LET of 87 keV/µm was 6.2 X 10^-11 ssb/Gy/Da (Fig. 1). The yields were found to be fairly similar for carbon ion particles with LETs of 122, 342 and 507 keV/µm. This value of around 6 X 10^-11 ssb/Gy/Da is comparable to that obtained after γ- irradiation. While the yield of prompt ssb does not alter with varying LET values from 87-507 keV/µm, the yield of prompt dsb appears to increase as the LET increases (Fig. 2). Consequently, the ratio of prompt ssb to dsb tends to decrease as the LET increases, indicating that strand breaks in close proximity, which would produce dsb’s, are more readily induced at higher LET.

Results of enzymatically induced ssb/dsb revealed that the amount of base lesions susceptible to cleavage by Nth and/or Fpg becomes less as the LET gets higher (Fig. 1 and Fig. 2). Although we cannot fully disregard the possibility that the number of damaged bases is actually reduced at higher LET, we favor the interpretation of base lesions being clustered and thus becoming less readily cleaved by the glycosylases. Inhibition of excision of damaged bases within a cluster has been indeed demonstrated using purified enzymes. It should be noted that the yields of prompt and enzymatically induced DNA damage after exposing to carbon ion particles with a LET of 87 keV/µm were lower than those of helium ion particles with a LET of 95 keV/µm. This result suggests that the LET is not fully sufficient a parameter to describe the nature of damage clustering and that rather the actual density of ionization should be taken into account.

In summary, our results indicate that DNA lesions in hydrated plasmids become more readily clustered on increasing ionizing density of the radiation.

References
3-32 Analysis of Cellular Effects and Local Damage Induced by High LET Heavy Ions

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Introduction

High linear energy transfer (LET) radiation, such as heavy ion beam, has greater biological effectiveness than low LET radiation. Higher biological effectiveness of high LET radiation is probably a consequence of local damage like clustered DNA damages that contain locally multiple oxidized bases, single strand breaks and double strand breaks (DSB), because it is considered that the clustered DNA damage is non-repairable or more difficult to repair than a lesion sparsely induced. On the other hand, mammalian cells efficiently detect and repair DNA damage. Especially, mammalian cells have evoked two pathway for repairing DSB, homologous recombination (HR) and non-homologous end joining (NHEJ). The major pathway in mammalian cells is NHEJ. Many proteins are involved in NHEJ which include Ku70/80 heterodimer, DNA-PKcs, XRCC4 and ligase IV. First, Ku bind to the ends of a DSB and DNA end binding by Ku initiates a cascade of molecular events that leads to joining of the broken DNA ends. In addition to DNA repair, Ku protein plays a role in multiple nuclear processes, such as chromosome maintenance and transcription regulation. However, much less is known about the yield and reparability of clustered DNA damages.Recently we suggest that the number of non-repairable DSB by Ku–dependent DSB repair system increases with increasing LET. In this study, to investigate non-repairable DSB by Ku–dependent DSB repair system we analyzed behavior of Ku protein to DNA damage induced by high LET heavy ions.

Material and Methods

We transfected pGFP-Ku80 into xrs-5 cells (mutated in Ku80). This cell was grown in MEM-alpha medium supplemented with 10 % serum and were incubated at 37 °C in a humidified atmosphere of 5 % carbon dioxide and 95% air. Accelerated ions of 220 MeV C 5+ (108 keV/μm), 460 MeV Ar 13+ (1610 keV/μm) were provided by the AVF cyclotron at TIARA JAEA-Takasaki. Cells were irradiated with ion beams. To evaluate DNA damage and NHEJ pathway induced by heavy ion beams, we carried out immunohistochemical detection of phosphorylated histone H2AX (γH2AX). Cells were attached on the CR-39 film that is a particle track detector. After post-irradiate incubation of 10-30 min, the cells were fixed by 1 % paraformaldehyde in PBS. Then, the cells were incubated with anti-γH2AX antibody for detecting DNA double strand break sites in situ. Thereafter, the cells were counter-stained with DAPI and the opposite side of the CR-39 film was etched with KOH-ethanol solution at 37 °C. Using a confocal microscope, we took images of the cells (γH2AX and GFP-Ku80) and the etched pits on the CR-39 film at a same field of view. By merging all images together, the position of hit-ions, γH2AX and GFP-Ku80 were simultaneously detected. Additionally, because Ku70/80 and DNA-PKcs formed complex in NHEJ pathway, in situ localization of DNA-PKcs was also analyzed.

Results and Discussion

In xrs5 -GFP-Ku80 cells, the site of γH2AX foci and the site of etched pits were coincident. In addition, GFP-Ku80 foci co-localized with those of γH2AX. These observations show that Ku80 proteins localize to DNA damages induced by ion beam irradiation, indicating that Ku80 protein can recognize and bind to DNA damages induced by heavy ion irradiation. The GFP-Ku80 foci were observed in the samples of 10 min to 30 min after C ion irradiation, and, at the same moment, DNA-PKcs also were localized to DNA damage sites. This observation suggests that the damage induced by C ion irradiation is repairable by Ku-dependent DNA repair pathway. On the contrary, the foci of GFP-Ku80 were observed in 10 min after Ar ion irradiation, but they disappeared in 20 min or later (Fig.1). In addition, co-localization of GFP-Ku80 and DNA-PKcs was not clear even in 10 min after Ar ion irradiation. These results indicate that Ku80 can bind to DNA damage induced by high LET heavy ion like Ar, but cannot form a complex with DNA-PKcs, resulting in failure of NHEJ repair of induced DNA damages.

Fig.1. Cell with co-localization signal of GFP-Ku80 and γH2AX after C ion and Ar ion irradiation.
低線量／低線量率放射線に対して生物が示す特異的な応答様式の一つに放射線誘発バイスタンダーエフェクトがある。我々は、日本原子力研究開発機構において開発された重イオンマイクロビーム細胞照射システム（細胞局部照射装置）を用いて、この放射線誘発バイスタンダーエフェクトの分子メカニズムを明らかにすることを計画した。

1. 実験方法

(1) 細胞：ヒト正常線維芽細胞（AG1522細胞）を用いた。

(2) 培養：開孔（径12 mm）35 mmディッシュの内面中央に20 mm四方のCR-39をパラフィンで固定したものを使用した。またCR-39の表面をcollagenコートして細胞を培養した。2×10³cells/mlの細胞懸濁液を5 µlにスポットし（1000 cells/colony）、15-20時間培養したものを照射実験に供した。

(3) 照射：Funayamaら２)の方法に従って、中央にスポットしたコロニーの細胞1個に5粒子の460 MeV ⁴₀Ar に照射した。また対照実験として460 MeV ⁴₀ArをHY1ポートにおいても照射した。

(4) 細胞の蛍光免疫染色：照射後、細胞を37℃で培養し、0.5および6時間後メタノールで固定し、リン酸化型H2AX（γH2AX）抗体およびリン酸化型NBS1（pNBS1）抗体を用いた蛍光抗体染色法により染色し、蛍光顕微鏡で観察した３)。標的細胞を含む正方形枠（250 x 250 µm；C内、そこから右方向に設定した）の正方形枠（250 x 250 µm；AおよびB）の内部、および遠隔に存在するサテライトコロニーの中心部の正方形枠（250 x 250 µm；S）内のフォーカス形成頻度を測定した。

2. 結果および考察

(1) 標的細胞でのγH2AXおよびpNBS1のフォーカス形成：Ar線マイクロビームの照射により、照射された細胞においてγH2AXおよびpNBS1のフォーカス形成を認められ、それは同所局在していた。

(2) バイスタンダー細胞におけるγH2AXおよびpNBS1のフォーカス形成：Ar線マイクロビーム照射により、バイスタンダー細胞においてもγH2AXおよびpNBS1のフォーカス形成が認められ、それらは同所局在していた。従って標的細胞から分泌される可溶性低分子物質（バイスタンダー因子）によりバイスタンダー細胞にDNA二本鎖切断が誘発されていることが示唆された。またバイスタンダー細胞におけるこれらのフォーカス形成は標的細胞から離れるに従ってその頻度は減少した。

(3) バイスタンダー因子の種類に依存したバイスタンダー効果の発現：Ar線マイクロビーム照射により、バイスタンダー細胞におけるγH2AXおよびpNBS1のフォーカス形成がDimethylsulfoxide（DMSO, 0.1%）、Lindane（40 µM）、Aminoguanidine（AG, 10µM）およびcarboxy-PTIO（c-PTIO, 10 µM）によって抑制された。DMSOはROSの消去剤、Lindaneは細胞内のギャップ結合の阻害剤、AGはnitric oxide (NO)ラジカル合成酵素の阻害剤、c-PTIOはNOラジカルの消去剤である。DMSOによる抑制は、CおよびAにおいて顕著であり、それ以外の部位では抑制効果は認められなかった。Lindaneによる抑制効果はCにおいてのみ顕著であった。AGおよびc-PTIOによる抑制効果は、A、BおよびSにおいて認められ、特にSにおいて最も顕著であった。

以上のようにギャップ結合を移行しているバイスタンダー因子および標的細胞近隣の細胞に培地中を介して作用しているバイスタンダー因子は活性酸素種（ROS）であることが示唆された。一方、遠隔的に作用しているバイスタンダー因子は活性窒素素種（RNS）、特にNOラジカルであることが示唆された。

References

3) A. Takahashi, et al., Cancer Res. 64, 8839 (2004).
Exposure of Normal Human Diploid Fibroblasts to Heavy Ions Facilitates Differentiation in Their Progeny

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Radiation-induced genomic instability encompasses a variety of delayed effects that occur in the progeny of survivors multiple generations after the initial insult. LET dependence of these effects is incompletely characterized; however, we have previously found that delayed reductions in clonogenicity can be most pronounced at LET of 108 keV/µm. To gain insight into potential cellular mechanisms underpinning LET-dependent delayed loss of clonogenicity, we investigated morphological changes in colonies arising from normal human diploid fibroblasts exposed to 60Co γ-rays (0.2 keV/µm) or carbon ions (18.3 MeV/amu, 108 keV/µm). Carbon ions were fourfold more effective at inactivating cellular clonogenic potential and produced more abortive colonies containing reduced number of cells per colony than γ-rays. Second, colonies were assessed for clonal morphotypic heterogeneity. The yield of differentiated cells was elevated in a dose- and LET-dependent fashion in clonogenic colonies, while differentiated cells dominated to a comparable extent irrespective of irradiation in abortive colonies. The incidence of giant or multinucleated cells was also increased but much less frequent than that of differentiated cells. All together, our results indicate that carbon ions facilitate differentiation more effectively than γ-rays. Accelerated differentiation may account for dose- and LET-dependent delayed loss of clonogenicity in normal human diploid cells, and could be a defensive mechanism that averts further expansion of aberrant cells.

References

γ線の10%生存線量に対する炭素線のRBEは3.9であった。線量とLETに依存して、コロニーあたりの細胞数は減少し、特に2-7細胞のコロニーの割合が著しく増加した。形態変化を起こした細胞の割合は、50細胞以上のコロニーでは線量とLETに依存して増加したが、50細胞未満のコロニーでは照射の有無に依らずほとんど一定であった。コロニー内には巨細胞や多核化細胞も認められ、照射された細胞の変化は、分化した細胞、巨細胞、多核化細胞のいずれもコロニー形成能を喪失していると考えられる。照射子孫細胞への形態変化の誘導は、2次コロニー形成能喪失の機序である可能性とともに、異常を蓄積した細胞の増殖を抑制するための防御機序である可能性が示唆された。また、分化した細胞の誘発頻度が線量依存的に増加したことから、DNA損傷が一因となり分化の誘導を促進するこが考えられる。放射線照射後に観察されるクロマチンの構造変化などに由来して形成されるゲノム上の潜在的なDNA損傷が、照射子孫細胞に遅延的に誘発されると考えられている。このことから、照射細胞に誘発されるDNA損傷が起因となって子孫細胞に遅延的に誘発されるDNA損傷のチェックポイント活性化することにより遺伝的安定性の形質が永続的に発現すると考えられる。今後は、重粒子線誘発遺伝的不安定性の分子機構を明らかにしていきたい。
3-35 Radiation Induced Apoptosis in *Drosophila* Cells

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Extracellular stresses including radiation were reported to increase intracellular ceramide concentration in mammalian cells. Cell-permeable ceramide, such as C2 ceramide which has shortened fatty-acyl chain, was able to induce apoptosis in many cell lines. These results suggested that ceramide acts as a signaling molecule in radiation-induced apoptosis of mammalian cells. Ceramide is generated either by de novo synthesis or sphingomyelin degradation catalyzed by sphingomyelinases. The sphingomyelin degradation pathway was reported that the major source for generating ceramide in radiation-induced apoptosis. There are three classes of sphingomyelinase based on their optimal pH, acid, neutral. Both acid sphingomyelinase (aSMase) and neutral sphingomyelinase were reported to be activated during apoptosis. The mechanisms that control radiation-induced activation of SMases are not well defined. It has been shown that key components in apoptotic pathway are well conserved between *Drosophila* and mammals.

We examined whether ceramide level is regulated during radiation-induced apoptosis in *Drosophila* Schneider line 2 cell (SL2). Three-fold increase in ceramide was observed 5 hours after UV-C irradiation. Addition of cell permeable ceramide, C2 ceramide, to SL2 cells increased Caspase 3/7 activity. Furthermore, overexpression of ceramidase that hydrolyzes ceramide caused reduction of Caspase 3/7 activity.

We carried out RNA interference experiment to clarify the ceramide-generating enzyme(s) during the apoptosis. Caspase 3/7 activity was decreased only by adding the ds RNA for neutral sphingomyelinase homolog. Other ds RNAs including acid sphingomyelinase homolog, and sphingolipid Δ4-desaturase had no significant effect. (Fig1)

These results suggested that ceramide have an important role in radiation-induced apoptosis and neutral sphingomyelinase homolog might affect the regulation of ceramide synthesis.

Fig 1. Effects of various dsRNAs for sphingomyeline-related genes on the hea

Double strand (ds) RNA was added to SL2 and incubated for 3 days to allow for turnover of the protein. To induce apoptosis cells were irradiated with heavy ion. DEVD-luciferin and luciferase was added to the cell lysate after 55 hours of irradiation, followed by measurement of Caspase 3/7 activity with luminometer. Filled bar: irradiated with 40 Gy of C5+ ion, Gray bar: no irradiation, Ark: Apaf-1 related killer, DES: sphingolipid Δ4-desaturase, aSMase1: acid sphingomyelinase homolog-1, aSMase3: acid sphingomyelinase homolog-3, EGFP: enhanced green fluorescent protein
3-36 Pathological and Immunohistochemical Findings of a Human Glioblastoma Cell Line (CGNH-89) after X-ray and Heavy-ion Beam Exposure

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Purpose
It has not been elucidated whether heavy-ion beam irradiation modulates the biology of glioblastoma at the cellular level. Our project is designed to investigate whether heavy-ion beam exposure is effective against glioblastoma cells. We examined a human glioblastoma cell line (CGNH89 1) by morphological and immunohistochemical techniques after X-ray or heavy-ion beam exposure.

Method
For X-ray and heavy-ion beam exposure, CGNH89 cells were divided into 3 groups; control, 5 Gy and 10 Gy exposure. For each group, cells were fixed at the same time-points. Hematoxylin-Eosin (HE) and immunohistochemical staining were done in formalin-fixed cells. For electron microscopy, cells were fixed in fixative (1 % glutaraldehyde, 10% formaldehyde). These samples were postfixed with osmium and embedded in Quetol 812. Ultrathin sections were examined under a transmission electron microscope.

Results and Discussion
After the exposure, both living cells and mitotic cells decreased in cell number. In the HE staining, nuclear condensation and apoptotic bodies increased after exposure. MIB-1 labeling index decreased in the early stage after exposure, but increased in later stages. It is suggested that cell cycle arrest and induction of apoptosis was induced by X-ray and heavy ion-beam exposure in CGNH89 cells. Ultrastructurally, the nuclei and cytoplasm after exposure showed a tendency to increase in size. Cellular swelling seems to depend on the swelling of mitochondria, increase of cellular organelles, cytoskeleton, and secondary lysosome (Fig.1,2). However we did not find out either apoptotic body or necrotic change. We conclude that X-ray and heavy ion-beam exposure might trigger the same morphological changes in the surviving CGNH-89 cells.

Reference
Bystander Killing of Human Lung Cancer Cells by Heavy Charged Particle Irradiation

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We have here examined clonogenic survival of confluent human lung cancer A549 cells exposed to X-rays or 220 MeV $^{12}$C$^{5+}$ charged particles. Targeted exposure of 0.001-0.005 % of cells within the confluent population were resulted in 7-14 % reduction of surviving fraction. However, this reduction was diminished when cells were treated with the inhibitor of gap junction. These results suggest that gap junctional intercellular communication play s an essential role in heavy charged particle irradiation-induced bystander effects.

1. はじめに
これまでのわれわれの検討から、ブロードビームを用いた実験で$^{12}$C$^{5+}$の細胞生存曲線は直線的であったが、X線および$^{2}$C$^{6+}$のD$_{0}$（37 %生存線量）／D$_{10}$（10 %生存線量）はそれぞれ1.59 Gy/4.00 Gy、1.23 Gy/2.39 Gyであった。この重イオンビーム照射による細胞死の機序にバイスタンダー効果が関与していることが知られているが、本研究では細胞間のギャップ結合に注目し、重イオンビーム照射後の細胞生存率について検討を行った。

2. 実験方法
細胞はヒト肺癌由来の細胞株A549を用いた（p53 statusは野生型）。重イオン照射は原子力機構TIARAのマイクロビームにて行った（220 MeV$^{12}$C、108 keV/µm）。照射後の細胞生存率はコロニー形成法で検討した。細胞を高密度（コンフルエント）と低密度に培養した状態で照射するため、照射2日前に底面に固定飛跡検出材（CR-39）を貼付した35mmのグラスディッシュ上に細胞を準備した（高密度群では5x10$^5$個、低密度群では5x10$^4$個）。標的細胞はディッシュ中の1-25個の細胞をランダムに選択し、照射するイオンの数はひとつの細胞あたり1-10個とした。照射後は60 mmのディッシュに至適個数の細胞をtriplicateでまきなおし、14日後に細胞を固定・染色した後、コロニー数をカウントし、非照射細胞の生存率を対照とした。また、高密度に培養した細胞ではギャップ結合の促進剤（+cAMP）を加え、バイスタンダー効果を検討した。

3. 結果および考察
コンフルエントな状態の細胞のうち選択的に1つの細胞にのみイオンを照射した際の細胞生存率は非照射細胞とはほぼ同様であった。照射細胞数をディッシュ内の5-25個、また各細胞への照射イオン数を5-10個とすると照射していない細胞の生存率に比較して7-14 %低下した（P<0.05：データの提示なし）。一方、低密度に細胞を培養し、細胞間のコンタクトをなくした状態でディッシュ内の細胞を選択的に照射したときの細胞生存率は非照射細胞と同様であった（データの提示なし）。コンフルエントに培養した細胞でギャップ結合を促進（+cAMP）するとバイスタンダー効果が促進される傾向にあった（p=0.07）。ギャップ結合を阻害（Lindane）するとバイスタンダー効果が有意に抑制された（P<0.05、Fig.1）。

本実験ではディッシュ全体の細胞のうち0.001-0.005 %という極少数の細胞を重イオンビームで照射した時、ディッシュ全体の細胞生存率が有意に低下する結果が得られたが、細胞間のコンタクトをなくした状態で照射するとその現象は認められなかった。またギャップ結合阻害剤を細胞に接触させることにより照射後の細胞生存率低下は抑制された。これらより重イオン照射による細胞死に照射細胞から非照射細胞へのバイスタンダー効果が関与しており、また、そのシグナル伝達経路にギャップ結合が深く関与していることが示唆された。

Fig.1 Relative surviving fraction of confluent density-inhibited culture of A549 cells where 25cells were irradiated with 5 carbon ions with or without treatment with Lindane or cAMP. The surviving fraction of the nonirradiated A549 control was normalized as 1. The plating efficiency of control was 46-57 %.
3-38 Effects of Heavy-ion Irradiation on the Expression of Cellular and Viral Genes

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Introduction
Heavy-ion irradiation, a new tool for cancer therapy and radiation biology, is expected to cause different responses on human cells from those by X-rays. The expression of some cellular genes is possibly induced by heavy-ion radiation. The aim of this study is to identify such genes using the susceptibility of human cells to human immunodeficiency virus type-1 (HIV-1) as an indicator. We have found that the susceptibility of human cells to HIV-1 is enhanced by heavy-ion irradiation. This enhancement was not detected in cells irradiated with X-rays. The mRNA expression of various cellular factors that are involved in HIV-1 replication, DNA damage or DNA repair were examined. Effects of various drugs on the mRNA expression of cellular factors were determined.

1. Materials and Methods
Human glioma cells transduced with CD4 and CCR5 genes, NP-2/CD4/CCR5 1), and NP-2/CD4/CCR5 cells transduced with green fluorescence protein (GFP) gene driven by HIV-1 LTR, NP-2/CD4/CCR5/LTR-GFP, were used in this study. NP-2/CD4/CCR5 cells were irradiated with carbon (C) ion or X-ray at different doses (0.5, 1.0, 2.0, and 4.0 Gy). Then, NP-2/CD4/CCR5 cells were exposed to a CCR5-using (i.e. R5-tropic) HIV-1 strain, Ba-L, 48 hour after irradiation. The susceptibility of NP-2/CD4/CCR5 cells to HIV-1 was determined by PCR to detect reverse-transcribed env DNA 24 and 48 hour after viral infection. NP-2/CD4/CCR5 cells treated with drugs.

2. Results and discussion
An amount of the reverse-transcribed env DNA was increased in NP-2/CD4/CCR5 cells infected with HIV-1 after heavy-ion irradiation at 0.5 and 1.0 Gy, suggesting that the susceptibility of the cells were enhanced by the radiation (Fig. 1). However, enhancement of the HIV-1 susceptibility could not be detected in NP-2/CD4/CCR5 cells irradiated with heavy-ion at 2.0 and 4.0 Gy. An amount of HIV-1 bound on NP-2/CD4/CCR5 cells irradiated with heavy-ion at different doses was similar, suggesting that the irradiation has little effect on the efficiency of HIV-1 binding on the cell surface. The enhancing effects of heavy-ion irradiation on the susceptibility of NP-2/CD4/CCR5 cells to HIV-1 may be caused by increased incorporation or genomic reverse-transcription of HIV-1.
Expression of CD133 (Prominin-1) and NF-kB mRNA was detected to be increased in NP-2/CD4/CCR5 cells irradiated with heavy-ion. Expression of histone deacetylase, Ku80, and poly-ADP ribose polymerase mRNA was decreased in NP-2/CD4/CCR5 cells irradiated with heavy-ion. However, a relationship between the enhancing effect of heavy-ion irradiation on the susceptibility of NP-2/CD4/CCR5 cells to HIV-1 and the altered expression levels of these cellular factors has not been elucidated well.

The susceptibility of NP-2/CD4/CCR5 cells to HIV-1 was upregulated when the cells were pretreated with 5-azacytidine, neuraminidase or Wortmannin, suggesting that chromosomal demethylation, inhibition of PI3-kinase, or modification of glycosylation of cell membrane factors may have a role in the enhancing effects of heavy-ion irradiation on the susceptibility of NP-2/CD4/CCR5 cells to HIV-1.

Effects of heavy-ion irradiation on expression and activities of other cellular factors should be analyzed to elucidate its biological effects on human cells.

Fig. 1. The susceptibilities of NP-2/CD4/CCR5 cells to HIV-1 after heavy-ion irradiation. (A) Effects of heavy-ion irradiation on binding of HIV-1 to cells. NP-2/CD4/CCR5 cells were irradiated with heavy-ion at different doses and exposed to HIV-1 Ba-L strain for one hour at 4℃. HIV-1 bound on the cells was detected by RT-PCR. Expression of GAPDH mRNA was detected by RT-PCR as controls. (B) Reverse-transcription of HIV-1 in irradiated NP-2/CD4/CCR5 cells. Reverse-transcribed HIV-1 DNA was detected by PCR after 24 and 48 hour after viral exposure.

Reference
Morphological Study on the Effects of Heavy Ion Irradiations to the Isolated Muscle Fibers of SJL Mice

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Muscular dystrophy is a hereditary disease characterized by progressive muscle wasting and degeneration. Although many genes affected by the disease have been elucidated, there remains a wide gap between the mutations of the genes and the development of the disease. We are trying to use irradiations of heavy ion beams to skeletal muscle cells to understand the pathophysiology of the disease.

In a major subgroup of muscular dystrophy, products of the affected genes are localized in or around the plasma membrane. Their absence is supposed to cause the fragility of the membrane, which leads to the degeneration of the muscle fibers. In a previous study1,2), we found that the irradiation of heavy ion microbeams on isolated single fibers of the skeletal muscle caused the damage of the sarcolemma, which induced irregular protrusions and invaginations of the plasma membrane, in addition to the changes of the cytoplasmic structures such as disarrangement of the myofilaments, enlargement of the cisternae of endoplasmic reticula containing flocculent matter and increase of autophagy.

Here, we report the results of heavy ion irradiation to the isolated single fibers of SJL mice, which have a mutation in dysferlin gene and are the model animal of limb-girdle muscular dystrophy type 2B and Miyoshi muscular dystrophy.

Single fibers of SJL mice were isolated by collagenase treatment and plated on CR-39 plastic cover slips. Specimens were irradiated with either microbeam (^{40}Ar: 11.2 MeV/amu or ^{20}Ne: 12.8 MeV/amu) or broadbeam (^{40}Ar: 460 MeV or ^{20}Ne: 260 MeV, total 1-10 Gy). After irradiation, the muscle fibers were fixed at various time points and processed for electron microscopy.

The irradiated fibers of SJL mice showed the same change as those found in control ICR mice (Fig. 1), namely irregular protrusions and invaginations of plasma membrane, disarrangement of thick filaments, distended cisternae of endoplasmic reticula containing flocculent matter, and increase of autophagy. In addition, vesicles of unknown origin and autophagic vacuoles intermingled with disarranged thick filaments were observed in some areas under the sarcolemma (Fig. 2). These vesicles may be concerned with the disturbed turnover of the membrane system, as dysferlin, which is absent in SJL mice, is proposed to be involved in the event.

References
Relative Biological Effectiveness of Carbon Beams on Cell Neurons; Approach Using Growth Cone Collapse Assay

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The relative biological effectiveness (RBE) on neurons are believed to be difficult to be evaluated due to the complexity for culturing them alone, which has meant that their radio-sensitivity has not been well investigated. The growth cone collapse (GCC) assay has been reported as a useful means of quantifying the effects of various factors on cultured explants of nervous tissue $^{1-2)}$. Here, we used the GCC assay to determine the RBE of carbon beams to X-rays on the cell neurons.

Dorsal root ganglia (DRG) and sympathetic ganglion chains (SYMP) were isolated from day-16 (mature) and day-8 (immature) chick embryos and cultured for 20 h $^{3)}$. Thereafter, neurons were exposed to graded doses of X-rays, or high-LET $^{12}$C ions (18.3 MeV/amu, 108 keV/μm). Morphological changes of the neurons were examined quantitatively by GCC assay. Apoptosis induction was examined using TUNEL assay (Fig. 1).

Carbon beams induced GCC and neurite destruction in a time and dose-dependent manner (Fig. 2). Day-8 neurons were more radiosensitive than day-16 neurons (p=0.01). At 12 h post-irradiation, 20 Gy carbon beams and 30 Gy X-rays induced about 65% and 25% apoptosis, respectively. The simple regression analysis revealed that the carbon beams RBE at day-8 DRG and SYMP using the GCC data were 4.6 and 4.2, respectively. Whilst, at day-16 DRG and SYMP were 3.4 and 3.3, respectively. However, the RBE at day-8 DRG and SYMP for apoptosis induction was 4.1 and 3.7, respectively, whereas that at day-16 DRG and SYMP was 4.2 and 3.7 respectively. The carbon beams were 3.3-4.6-fold more effective than X-rays for inducing GCC and apoptosis on neurons. Our implication is that attention should be considered when estimating the therapeutic dosage of carbon beams clinically. GCC assay could be beneficial in in vitro radiobiological investigations.

References
3-41 Cell-killing Effect in Bystander Cells Induced by Carbon-ion Microbeams

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A central paradigm in radiation biology has been that only cells “hit” by a track of radiation would be affected to induce radiobiological consequences, and cells “not hit” should not be. This paradigm is the basis for the current system for risk estimation of radiobiological effects. Also, the risk of radiation-induced cancer after high and moderate doses is relatively well known, based on the data from detailed epidemiological studies of the Japanese atomic bomb survivors in Hiroshima and Nagasaki1). However, it recently has been challenged by so called non-targeted effects, such as genomic instability, adaptive response and bystander effect, and such radiation-induced non-targeted effects may have important implications for risk evaluation of low dose / low dose rate radiations.

This year, we have begun to investigate cellular bystander responses in normal human fibroblasts induced by carbon-ion microbeams. Carbon-ion microbeams (12C5+, 220 MeV) were generated with the HZ1 port at TIARA. Approximately 6 x 10^5 exponentially growing cells were inoculated into each of microbeam dish, which was constructed with an aluminum ring of 36 mm diameter and attached 7.5 µm-thick polyimide film on the bottom of the ring, 2 days before irradiation. One day after plating, half of the dishes were treated with a specific inhibitor of gap-junction mediated cell-cell communication (40 µM of γ-isomer of hexachloro-cyclohexane). At the irradiation period, cultures were confluent. Cell cycle distribution of the confluent cultures were analyzed using a flow cytometry and around 95% of the cells were G_1- or G_0-phase. Irradiation was carried out using a 256 (16 x 16)-cross-stripe method (Fig.1). Microbeams of 20 µm in diameter were irradiated in each point with 8 delivered ions.

Figure 2 showed cell-killing effect, which was detected with a colony-forming assay as a reproductive cell death, in microbeam-irradiated dishes (IR) and microbeam-irradiated dishes with a specific inhibitor of gap-junction mediated cell-cell communication (IR+L). The percent cell survival in microbeam-irradiated dishes was ranging from 83% to 94%, while from 97% to 100% in microbeam-irradiated dishes with a specific inhibitor of gap-junction mediated cell-cell communication. In our microbeam-irradiation method, we estimated that the percent of C-ion direct hit cells was ranging from 0.08% to 0.2% of all cells in the dish. The results showed a higher cell-killing effect in the microbeam-irradiated cell population than that with a specific inhibitor of gap-junction mediated cell-cell communication. Our studies provide clear evidence that microbeam-irradiated cells can induce a bystander lethal response in neighboring cells, which are not directly hit by C ions and gap-junction mediated cell-cell communication play an important role in the bystander effect.

Reference
3-42 Ion Beam Irradiation Has Different Influences on Glutathione Peroxidase of Cultured Human Retinal Vascular Endothelial Cells Exposed to L-dopa among $^{20}$Ne, $^{12}$C and $^{4}$He

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It is well-known that the damage of retinal endothelial (RE) cells is one of causes as pathogenesis in senile macular degeneration and diabetic retinopathy. Glutathione peroxidase (GPX), a selenium-dependent and lipid peroxide-scavenging enzyme that effectively reduces lipid peroxides with the concomitant oxidation of glutathione is distributed in mitochondria 1). Faucher et al. measured the expression of two bcl-2 family members, bax and bcl-2, in a human endothelial like cell-line overexpressing the organic hydroperoxide-scavenging enzyme GPX, in the absence of any apoptotic/oxidant stimulus, and showed that overexpressing an antioxidant gene such as GPX in endothelial cells is able to change the basal mRNA and protein bax levels without affecting those of p53 and bcl-2. This phenomenon could be useful to antiatherogenic therapies which use antioxidants with the aim of protecting the vascular wall against oxidative stress injury 2). The irradiation is known to result in a dose-dependent decline in the activities of GPX in the skin of mice 3). The gamma ray ($^{60}$Co) causes ionization uniformly in the whole irradiated tissue. We found out that high dose of gamma irradiation prevented programmed cell death regulated by p53 and bcl-2 genes in the ciliary body dissected from living body, and was useful for preservation of organ after culture by the protective influence of inflammatory reaction 4). Deficiency of L-dopa causes degeneration of the substantia nigra in the brain and L-dopa is used in the treatment of Parkinson’s disease. We proved that L-dopa produced NO and superoxide, and had the cytotoxic effects on the retinal pigment epithelial cells 5). L-dopa injected into the vitreous of the rats dilated the vena in the ciliary body 6). We wondered if RE cells could be exposed to oxidative stress by administered L-dopa. We already reported that oxidative stress such as hyperoxia 7) augmented the cytotoxicity of the aortic endothelial cells by L-dopa. We investigated how L-dopa influenced GPX in mitochondria and the cytoplasm of the RE cells that prevented from damage of phospholipid of cell membrane. We applied the ion beams as the induction of the oxidative stress that influenced GPX against in the RE cells, and measured the expression GPX with LightCycler system as a real-time-reverse-transcriptase polymerase chain reaction (RT-PCR).

Established human RE cells in vitro incubated with L-dopa (250 µM) for 2 hrs were exposed to an ionization radiation that is induced by acceleration of the ionizing atom of 350 MeV $^{20}$Ne, 220 MeV $^{12}$C, and 50 MeV $^{4}$He. We obtained the RE cells after 0, 4, 8, 24 hr of the irradiation and extracted total cellular RNA and cDNA was synthesized. We used the Primer3 website to design the primers for RT-PCR amplification of the cDNA of GPX and 18S RNA. The reactions were carried out at the following temperature: 95 °C, for denaturation; 60 °C, for annealing; and 72 °C, for extension for 17–27 cycles. After mixing the cDNA, primer, and SYBR green, the expression of 18S RNA and GPX was measured using the LightCycler system. The technology of this system is extremely innovative and enables rapid and simultaneous evaluation PCR experiments. Fluorometric analysis of the formed PCR products was performed as a real-time measurement either continuously or at specifically defined time points during each PCR cycle.

Exposure to L-dopa inhibited the expression of GPX in RE cells. Ion beam irradiations both of $^{4}$He and $^{12}$C decreased more remarkably than $^{20}$Ne. The expression of GPX in RE cells incubated with L-dopa decreased significantly after 8 hrs of exposure to $^{4}$He, and after 4 and 24 hrs of exposure to $^{12}$C. On the contrary, ion beam irradiation of $^{20}$Ne increased the expression of GPX in RE cells incubated with L-dopa after 4 hrs of the exposure of the irradiation significantly. We considered the different accumulation of the energy irradiated at a point by various ions could be concerned with the effects on the expression GPX in RE cells incubated with L-dopa.

References
3-43 Analysis of Lethal Effect Induced by Ion Beams in Canine Spontaneous Tumor Cells

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Due to the recent understanding of animal diseases and preventive medication including vaccination and premedication, companion animals can live much longer than before. These changes made it possible to reduce severe infectious diseases, and then made diseases of companion animals shift from infectious diseases to the aging-related metabolic disorders and neoplasm (cancer) formation. Radiotherapy is considered to be one of the good choices for the medical treatment against cancer. In human medicine, new radiotherapy such as high-LET heavy-ion beam radiotherapy has been introduced, because of its higher lethal effect than that of low LET radiation such as X-rays and $\gamma$-rays. However, in veterinarian medicine, there is still limited data on X-ray and $\gamma$-ray radiotherapy to dogs and cats, and needless to say, no study on ion beam radiotherapy. Therefore, to establish a proper protocol for radiotherapy on the animals, we investigated the fundamental aspects of the radiation effects on cancer cells of companion animals.

Even in the human medicine, the protocol for heavy ion radiotherapy still cannot be say well optimized, and the investigation for its establishment is still undergone. Of course, the dose per fraction of heavy ion therapy may become lower than that of X-ray therapy, because of its higher lethal effect. However the effects of low dose radiation has only been estimated by extrapolation from the data obtained by the higher dose radiation and not considered a concept of low dose specific biological responses such as adaptive response, radiation induced bystander effects, and low dose radio-hypersensitivity. Because the effect of those phenomenons cannot be estimated by extrapolation from the data obtained by high dose exposure, we also investigated lethal effects induced by low dose/dose-rate radiation that is considered to be important on heavy ion radiotherapy.

Cell lines used for this experiment were melanoma cell from spontaneously generated canine tumor. The cells were irradiated with 20 MeV H\textsuperscript{+} ions (2.7 keV/\mu m), 220 MeV $^{12}$C\textsuperscript{5+} ions (108 keV/\mu m) or X-rays at a dose rate of 0.8 Gy/min at a room temperature and replated on tissue culture dishes at a given density to measure the rates of the surviving cells. About 8 days after the irradiation, colonies of the cells were stained and counted. The survival rates against the given doses were plotted and fitted to a linear-quadratic model.

The survival curves of carbon ion irradiation showed almost linear in logarithmic scale and had no shoulder, but the curves of proton and X-ray irradiation showed to have shoulders. The lethal effect was highest by carbon ion irradiation among three types of radiation. The relative biological effectiveness (RBE) of proton and carbon ions calculated from the dose required to give 10 \% of survival rate was 1.32 and 2.82, respectively. These results indicate that the lethal effect increase a with LET value of ion beams.

The low dose survival rates measured by proton and X-ray irradiation showed good agreement to the extrapolated curve from the data of high dose irradiation. On the other hand, low dose survival rates by carbon ion irradiation were less than the estimated value from extrapolated curve of the high dose irradiation (Fig. 1). This result indicates that the lethal effect by low dose carbon ion irradiation is higher than estimated effect based on higher dose irradiation data. Additionally, we observed the treatment with DMSO, a radical scavenger, after low dose carbon ion irradiation restored the rate of survival to the value that is extrapolated from high dose irradiation (Fig. 2). This result indicates there is some participation of radicals in killing effects of low-dose carbon irradiation. In conclusion, the application of carbon ion radiotherapy to malignant melanoma therapy on companion animals would lead high therapeutic effects.

![Fig. 1 Rate of survival of canine melanoma cell lines after low dose exposure. Lines of survival curves were extrapolated from 1-8 Gy irradiation data.](image1)

![Fig. 2 Comparison of lethal effect by treatment with or without DMSO after low dose carbon ion irradiation. Dashed line was extrapolated from 1-4 Gy irradiation data.](image2)
3-44 Food-NaCl Associative Learning in Response of C. elegans to Gamma-ray Irradiation

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Learning impairments following ionizing irradiation as well as DNA damages is an important potential risk for astronauts1). We used Caenorhabditis elegans as a neuronal model organism, and previously reported that ionizing radiation affected food-NaCl associative learning behavior only at the transition stage of learning conditioning, which induced additional decreases in chemotaxis to NaCl immediately after irradiation. In the present study, we investigated the effects of gamma-ray irradiation on the time-course and chemotaxis to benzaldehyde.

We used well-fed adults grown on the plate containing 10 ml of the nematode growth medium agar3) and spread with the E. coli at 20 °C in all experiments. Using the method of Saeki et al.2) with some modifications, we evaluated the chemotaxis to NaCl. Animals move freely for 15 min on the 6-cm diameter assay plate with a gradient of NaCl concentration. When animals reach to the test spot or control spot, they were paralyzed by NaN₃. The test spot is the place with highest concentration (100 mM) of NaCl in the plate. The control spot is 4 cm away from the test spot. The chemotaxis index (CI) was calculated as the following equation; CI = { (number of animals within 1.5 cm diameter of the test spot) – (number of animals within 1.5 cm diameter of the control spot) } / (total number of animals on the assay plate). For the benzaldehyde assay, we used the 10-cm plates, in which 1 µl of ethanol or 0.5 % vol/vol benzaldehyde in ethanol was spotted at the control or test spot, respectively. The test spot was 6 cm away from the control spot, and NaN₃ was spotted to the both.

To test the effects of gamma-rays on the time-course, animals on the NaCl-conditioning plates (food-/NaCl+) were exposed to gamma-rays from ⁶⁰Co sources, Japan Atomic Energy Agency (JAEA). We measured chemotaxis at the time points of immediately after irradiation, 1, 3, 5 h after irradiation. NaCl-conditioning was continued during the post irradiation. As shown in Fig. 1, animals exposed to gamma-rays showed significant decreases in chemotaxis immediately after irradiation and complicated responses on post-irradiation. Animals exposed to the dose of 118 or 332 Gy showed almost the same level in CI 5 h after irradiation, whereas the CI of animals exposed to the highest dose increased up to the about 0.4.

Gamma-ray irradiation during the stage of conditioning for associative learning induced the significant decrease in CI of C. elegans, and then did show the complicated responses. We had the question whether other functions of the nervous system were also affected by gamma-ray irradiation. Thus, we examined the chemotaxis to benzaldehyde of animals with same conditioning and exposure as the previous result (Fig. 1). The CIs for chemotaxis to benzaldehyde were 0.84 ± 0.04 (0 Gy), 0.82 ± 0.03 (481 ± 32 Gy) immediately after irradiation, and 0.81 ± 0.02 (0 Gy), 0.81 ± 0.03 (481 ± 32 Gy) 5 h after irradiation. There was no significant difference in CI. Thus, animals exposed to gamma-rays did show the normal chemotaxis to benzaldehyde, whereas chemotaxis to NaCl was affected at the transition stage of associative learning.

These results may suggest the specific effects of gamma-rays on functions of the nervous system in C. elegans.

Figure 1. Time course of CI in NaCl-conditioned and exposed animals during the post irradiation. The error bars indicate the standard error of mean (SEM) from 5 or more experiments.

References
3-45 Food-NaCl Associative Learning in Response of *C. elegans* to High-LET Carbon Ion Beam Irradiation

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Learning impairments following ionizing irradiation is an important potential risk for astronauts as well as DNA damages1). Using *Caenorhabditis elegans* as a neuronal model organism, we previously showed that ionizing radiation affected food-NaCl associative learning behavior only at the transition stage of learning conditioning, which induced additional decreases in chemotaxis to NaCl immediately after irradiation. In the present study, we investigated the effects of high-LET carbon ion beam irradiation on the ability of associative learning and chemotaxis to benzaldehyde.

Well-fed adults grown on the plate containing 10 ml of the nematode growth medium agar2) and spread with the *E. coli* at 20 °C were used in all experiments. Chemotaxis to NaCl was measured by the method of Saeki et al.3) with some modifications. Animals (*C. elegans*) move freely for 15 min on the 6 cm diameter assay plate with a gradient of NaCl concentration. When they reach to the test spot or control spot, they were paralyzed by NaN₃. The test spot is the place with highest concentration (100 mM) of NaCl in the plate, and the control spot is 4 cm away from the test spot. The chemotaxis index (CI) was calculated as the following equation; CI = (number of animals within 1.5 cm diameter of the test spot) – (number of animals within 1.5 cm diameter of the control spot) / (total number of animals on the assay plate). For the benzaldehyde assay, we used the 10 cm plates. 1 µl of ethanol or 0.5 % vol/vol benzaldehyde in ethanol was spotted at the control and test spots, respectively. The test spot was 6 cm away from the control spot, and NaN₃ was spotted to the both.

To test the effects of carbon ions on the ability of associative learning, animals on the mock-conditioning plates (food-/NaCl-) were exposed to ¹²C (220 MeV, 113.3 keV/µm) delivered from the azimuthally-varying-field cyclotron installed in the Takasaki Ion Accelerators for Advanced Radiation Application (TIARA) facility, Japan Atomic Energy Agency (JAEA). Immediately after irradiation, animals were transferred to the NaCl-conditioning plates (food-/NaCl+) and placed for 4 hours to perform the conditioning for the food-NaCl associative learning. Subsequently, the chemotaxis assays were carried out. As shown in Fig. 1, there was no significant difference in CI between exposed and non-exposed animals. Thus, the result suggests that exposure to carbon ions dose not induce the decrease of the ability of food NaCl associative learning in *C. elegans*. This result is in accordance with that of animals exposed to ⁶⁰Co gamma rays.

![](image1.png)

**Fig. 1.** Dose response of the ability of associative learning. The error bars indicate the standard error of mean (SEM) from 4 experiments.

We have been reported that carbon ion irradiation during the stage of conditioning for associative learning induce the significant decrease in CI of *C. elegans*. The finding raises the question whether other functions of the nervous system are also affected by carbon irradiation. Thus, we examined the chemotaxis to benzaldehyde of animals with same conditioning and exposure as the previous report. There was no significant difference in CI (Fig. 2). Thus, animals exposed to carbon-ions showed the normal chemotaxis to benzaldehyde, whereas chemotaxis to NaCl was affected at the transition stage of associative learning. These results suggest that the carbon ion irradiation did not affect all functions of the nervous system in *C. elegans*.

![](image2.png)

**Fig. 2.** Dose response of chemotaxis to benzaldehyde. The error bars indicate the standard error of mean (SEM) from 4 experiments.

**References**

3-46 Effect of Energetic Heavy-ion Irradiation on Gene Expression in Caenorhabditis elegans

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Heavy ion irradiation, which is more effective than γ irradiation in inducing DNA damage and gene mutation, has been used for cancer therapy to kill tumor cells. Masumura et al. 1) reported that carbon particles induce deletions, generally greater than 1,000 base pairs in size, whereas γ-rays induce base substitutions and deletions of less than 100 base pairs. Other different effects of heavy ion irradiation are not well understood. We, therefore, study the effect of heavy ion and γ irradiation on gene expression with DNA microarray analysis of entire genome in Caenorhabditis elegans.

As a model organism C. elegans is easy to handle and genetically well-characterized, and its development and life cycle are well understood. In addition, the transcriptional profiles of individual cells or tissues at different developmental stages have been previously reported2). Here, we compared the transcriptional profiles of nematodes that were exposed to either 12C5+ ion broad beam (220 MeV) delivered from the AVF cyclotron or 60Co γ-rays using the whole genome C. elegans GeneChip array (Affymetrix, Santa Clara, CA).

To prepare the RNA for microarray analysis, N2 bristol wild-type hermaphrodites were synchronously cultured on Escherichia coli OP-50 NGM plates at 20 °C. Young adult hermaphrodites (24 h after the L4 larval stage) were exposed to 100 Gy of 12C5+ ion beam and γ-rays, and were collected 4 h later. Total RNA in each 103 worms was isolated with the TRIzol Reagent (Invitrogen Corporation, Carlsbad, CA) and was used as a template for generating probes using the One-Cycle Target Labeling and Control Reagents package (Affymetrix, Santa Clara, CA). Transcriptional alterations between heavy ion irradiated samples, 100 Gy γ-irradiated samples and non-irradiated controls were measured. Following heavy ion irradiation, 297 transcripts (approximately 1.3 % of the total transcriptome) were upregulated 2 fold or greater (Fig. 1). 388 transcripts were reduced at least 2 fold (Fig. 1). On the contrary, 187 transcripts were upregulated 2 fold or greater and 358 transcripts were downregulated at least 2 fold after γ-ray irradiation (Fig. 1). Interestingly, there were only 51 (upregulated) and 54 (down regulated) transcripts that were similarly affected by both irradiations (Fig. 1).

We further characterized the transcripts which were upregulated following treatment according to gene function. The expression levels of genes related to apoptotic inducer containing BH3 domain, egl-1 and ced-13, and glutathione S-transferase genes, cdr-2, cdr-3, gst-5 and gst-6, were significantly upregulated by either irradiation. It suggests that apoptosis and oxidative stress are similarly induced by 100 Gy of 12C5+ ions and γ-rays.

In contrast to γ-irradiated worms, certain genes mediating response to endoplasmic reticulum (ER) stress were significantly and specifically induced in heavy ion-irradiated worms. These results suggest that the heavy ion irradiation may efficiently cause both DNA damage and ER stress. It has been known that heavy ion particle irradiation efficiently causes apoptotic cell death3, 4). All together, ER stress induced by heavy ion particle irradiation may be a direct trigger of apoptotic induction.

References
3-47 Radiation Tolerance Linked to Anhydrobiosis in Polypedilum vanderplanki

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The larvae of the sleeping chironomid, Polypedilum vanderplanki, are able to withstand almost complete desiccation during which they enter a state of suspended animation1); this phenomenon is known as anhydrobiosis3). The anhydrobiotic larva can tolerate various extreme conditions including high doses of radiation3,4). Since physiological status is drastically changed during entry into anhydrobiosis3), it is of great interest to know how the desiccating larvae gain radiation tolerance.

We first examined effects of high-LET radiation on 4 kinds of larvae (Fig. 1): normal hydrated larva (W-low), intermediates between the anhydrobiotic and normal hydrated state (W-mid), almost completely dehydrated (anhydrobiotic) larvae (D-high), and immediately-rehydrated larvae that are assumed to have a similar molecular profile to anhydrobiotic larvae (W-high)4). These samples were exposed to 1 to 4500 Gy of 50 MeV 4He (LET=16.2 keV/µm) ion beam delivered from the azimuthally- varying-field cyclotron at TIARA. Irradiated larvae were supplied with distilled water and diet. Larvae that moved their pharyngeal plunger were considered to be survivors.

At doses between 3500 and 4500 Gy, differences in radiation effects became pronounced, depending on the physiological state of the larvae (Fig. 2). For all doses studied, survival period after radiation was greatest in D-high, followed by W-high, W-mid, and finally W-low. This result indicated that radiation tolerance increased as anhydrobiosis progresses. We previously thought that tolerance against various stresses in P. vanderplanki larvae is achieved by a complete desiccation. However, radiation tolerance was increased even in hydrated larvae as well, suggesting the protection mechanisms different from what we have speculated in desiccated state.

It is possible that trehalose itself has radioprotective effects in hydrated larvae of P. vanderplanki3). However, high-LET radiation mainly causes harmful damage to DNA by direct action, which is not reduced by protective agents. Therefore, it could not be determined whether trehalose inhibits a direct action of high-LET radiation in P. vanderplanki. Alternatively, we assume that DNA repair capacity is increased in desiccating larvae5). We reexamined the EST (expresses sequence tag) database from desiccating P. vanderplanki and found that fragments annotated as antioxidation enzymes such as thioredoxin reductase and glutation peroxidase, and DNA repair enzymes such as DNA photolyase and TFIH, increased during dehydration. Full-length cDNA cloning awaits further studies.

Damage from radiation shares common traits with desiccation damage: e.g. oxidative stress and DNA break. Many anhydrobiots seem to cope with that challenge by antioxidative defense and DNA repair systems. Prompt repair of DNA upon rehydration can be achieved by provision of efficient repair systems during entry into anhydrobiosis. Therefore, the increased radiation tolerance in P. vanderplanki presented here is probably due to cross-protective effects caused by anhydrobiosis6).

References
Study on Regeneration Mechanism of the Heavy-ion Irradiated Hemopoietic Organ of the Silkworm, *Bombyx mori* -Timing of Degradation and Regeneration of the Irradiated Organ -

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Our previous studies elucidated that, in *Bombyx mori*, the hemopoietic organ disrupted by the heavy-ion irradiation, would regenerate at wandering stage. This observation is an interesting phenomenon to investigate the hemopoietic stem cells in *B. mori*. However, the detailed mechanisms for degradation or regeneration on the irradiated organs have been obscured. In this study, the morphological changes of the hemopoietic organs after irradiation with 100 Gy of carbon-ions at day 0 of 4th instar were observed. In addition, the stage-specific marker proteins were also identified by 2-D electrophoresis followed by peptide-mass fingerprinting (PMF).

The timings of degradation and regeneration of the organ irradiated at day 0 of 4th instar occurred at almost identical stages to those of the organs irradiated at day 3 of 4th instar. These results predict that the developmental condition in larva, including the hormonal condition, might influence the timing for the degradation and regeneration of the irradiated organ.

Two prominent protein spots were detected in the sample from the organs being disrupted (at day 2 of 5th instar) by 2D electrophoresis. Those two proteins, estimated as soluble alkaline phosphatase and eIF 2 alfa kinase by PMF, would be involved in the degradation process of the organ.
Influence of Local Irradiation with Heavy-ion Microbeam on the Incidence of Somatic Mutation Arising on the Larvae in Embryo and Yolk in the Egg of the Silkworm, *Bombyx mori*

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Irradiation of diapause eggs (heterozygote) with heavy ion beam induces somatic mutation appearing as white spots on the black integument during larval stage. The incidence of the somatic mutation was clearly estimated as individual level dependently on both dose and LET\(^1\). The sensitivity was the highest level during the first 2 days after resumption of embryonic developments in the eggs, which terminated diapause status. Low dose irradiation with 2 mGy could induce the mutation \(^2\).

It remains unclear whether the somatic mutation would be induced by direct hit of the heavy particles to embryo, or indirect hit to yolk surrounding the embryo by bystander effect. This leads us to examine the incidence of the somatic mutation by local hit of heavy ion particles to embryo or yolk in *Bombyx* eggs.

The eggs \((P^0/p)\) were kept at 25 °C for first 10 days after oviposition, and then at 5 °C for 70 days to terminate diapause. These diapause-terminated eggs were transferred again to 25 °C for resumption of embryonic development. Carbon ion beam (3.0 Gy and 6.0 Gy) was irradiated to the 3-day old eggs after the resumption, and locally to 20μm of the center (yolk) of the egg at 112 keV/μm and embryo at 119 keV/μm. After irradiation, the newly hatched larvae were reared on an artificial diet throughout all stages, and we counted the number of larvae with white spots on the black integument to examine the incidence of the somatic mutation.

For local irradiation, 10 eggs were lined up on vertical and horizontal line in 1 cm length each, namely 98 eggs were placed on 1 cm\(^2\) of plastic film. But we did not place eggs at left corner on the first upper line and at right corner on the 10th lowest line to set fix points on the coordinate. The location of abdominal and caudal portion of embryo in each egg was determined under microscope, and was input in PC. According to this data, carbon ion-beam was hit on the definite location on each egg.

We set up two controls, one is placed at Kyoto and the other is carried to TIARA to check the influence of transport.

The incidence of the somatic mutation was 5 % in control of Kyoto, and 12 % in TIARA (Fig. 1). However, the irradiations of 3 Gy and 6 Gy to the center of egg did not result in the significant incidence of mutation between Kyoto and TIARA (Fig. 1).

In contrast, irradiation of 3 or 6 Gy to abdomen near caudal portion of embryo induced 63 % and 80 % of somatic mutation, respectively (Fig. 1).

In addition, irradiation of carbon beam to head had the almost same level in the incidence of somatic mutation as the control (non-irradiation), but the irradiation of 5 Gy to abdomen of embryo induced 80 % of the incidence (Fig. 1). The white spots appeared near the location which was hit by ion-beam.

From these results, it is not said that somatic mutation was resulted by bystander effect.

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References
3-50 Functional Analysis of Root Elongation Zone in Hydrotropism in Arabidopsis Using Heavy-ion Microbeam

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Previous studies on root hydrotropism suggest the importance of auxin response at elongation zone, however, there has been no functional confirmation of these necessities in this response. To determine the responsible tissue for root hydrotropism, it is necessary to inactivate the cell at desired position. Laser ablation of a specific cell can be used for similar purpose, and in fact, it has been used to dissect the functional role of root cap cells in gravitropism. Although this cell ablation is a powerful technique for understanding the function of certain cells, it is still difficult to determine the role of elongation zone, since application of this method to this tissue leads the root to be snapped and become unsuitable to monitor the root curvature. To solve this problem, we used heavy-ion microbeam irradiation for nondestructive cell inactivation method. Although irradiation with charged particles induce activation of specific genes in mammalian cells, irradiation of heavy charged-particles with relatively high linear energy transfer is thought to cause cell inactivation through induction of DNA double strand breaks. Recently, a device for heavy-ion microbeam irradiation with high precision has been established, and used to determine the effect of irradiation on root gravitropism. However, the interpretation of the effect of irradiation had been ambiguous, because, to the best of our knowledge, no report has demonstrated that irradiation of heavy-ion beam inactivates cellular processes by monitoring gene expression of interest in plant cells. In this study, we investigated the positional effect of cell inactivation by heavy-ion microbeam irradiation with root hydrotropism, together with the confirmation whether irradiation of heavy-ion beam suppresses auxin-inducible gene expression in plant species.

First, we tested whether heavy-ion beam irradiation could inactivate the cell, by investigating the alteration of auxin-responsive gene expression by exogenously applied auxin. In the following experiments, we used cold-run seedlings (seedlings without irradiation) as control samples to exclude the possibility that the experimental procedure itself affects the gene expression levels and the phenomenon. When control Arabidopsis seedlings were treated with 10^{-6} M indole acetic acid (IAA), 43.3-fold increase in the abundance of mRNA for auxin-responsive gene, namely IAA5, was observed as compared with mock cultures. On the other hand, when Arabidopsis seedlings were systemically irradiated by 220 MeV $^{12}$C$^{5+}$ ion at a dose of 500 Gy, only 8.3-fold induction in the abundance of mRNA for IAA5 was observed, which is far less than control. This indicates that the heavy-ion beam irradiation causes the dull responsiveness to exogenously applied auxin.

Next, we monitored the positional effect of heavy-ion beam irradiation on root hydrotropic response by irradiating with 220 MeV $^{12}$C$^{5+}$ ion at a dose of 500 Gy. We chose 180 μm aperture for irradiation so as to irradiate the whole part of elongation zone proximal to root tip. Control Arabidopsis seedlings responded to water potential gradient and developed orthotropic curvature. When cells at elongation zone proximal to root tip were exposed to heavy-ion microbeam irradiation, hydrotropic response is significantly repressed especially at the early phase of the hydrotropic response (data not shown). Our present results indicate that elongation zone profoundly affects the development of hydrotropic curvature, probably involving de novo auxin-responsive gene expression.

Figure 1. Effect of heavy-ion beam irradiation on auxin responsiveness.

Total RNA extracted from either $^{12}$C$^{5+}$-irradiated or non-irradiated samples was subjected to quantitative reverse transcription PCR. The amount of IAA5 transcript was normalized with ACT2 transcript level, and expressed as relative value where the normalized IAA5 transcript level in auxin omitted samples was set as one. Open bars indicate non-irradiated samples, whereas grey bars indicate irradiated samples. Data are the means of three independent experiments ± SD. Note that statistically significant difference was observed between two samples as calculated using Student’s two-tailed t-test ($P < 0.05$).

References
Differences in Circadian Rhythms of Resistance to \(\gamma\)-rays and Heavy-ion Beam in *Euglena*

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The algal flagellate *Euglena gracilis* (Klebs Z) displays circadian rhythms in resistance to \(\gamma\)-rays and heavy-ion beams: they persist for several days under continuous darkness at a constant temperature, and the most resistant phases occur 2-4 h before subjective mid-days, at which circadian rhythms in UV resistance and anti-oxidative capacities become maximal. We report here how these rhythms are related to each other. First, irradiation temperatures influenced neither survival after exposure to UV, \(\gamma\)-rays, nor C ions, whereas the incubation at high temperatures (25\(^\circ\)C) after irradiation considerably lowered the survival particularly after \(\gamma\)-irradiation, but not after UV irradiation at all. Secondly, although the antioxidants \(\beta\)-carotene and dimethylsulfoxide (DMSO) increased the survival of the alga at the least resistant phase after UV-C and UV-B irradiation, respectively, to the level of the most resistant alga, none of these were not so effective for \(\gamma\)- or C-ion-irradiation. Instead, GSH, GSH-ethylester, l-cysteine and l-histidine increased the resistance to \(\gamma\)-irradiation, while the other set of antioxidants did so for the resistance to C-ions. Particularly interesting was that the antioxidants l-ascorbate, N-acetyl-l-cysteine, rutin, trolox increased the latter but not the former resistance. Finally, FITC-dextran was incorporated into the alga only after C-ion irradiation but not \(\gamma\) irradiation, suggesting that C-ions may perforate cell membranes to kill the alga.

概日リズムは恒常環境の下ではその生物に固有な周期性（約24時間；ユーグレナで26時間）を表すとともに、昼夜の周期環境に同調して環境適応する。ユーグレナ (*Euglena gracilis* Z) では、UVC耐性やUVB耐性、及び抗酸化能の全般（脂溶性and/or 水溶性の一重項酸素消去能・ヒドロキシルラジカル消去能）が主観的真昼に最大となるよう概日リズムによって制御され、真昼に最大化する光酸化ストレスに対する予知的な防衛機能を果たしている。

実験にはユーグレナを6～8万細胞/mLになるまで25\(^\circ\)C、6 klx 恒明（白色蛍光灯）で培養し、恒暗に移す。恒暗中の様々な時点で \(\gamma\)-線や220 MeV 12Cイオン（以下Cイオン）に対してもユーグレナは同様の概日リズムを示すが、その最大耐性は主観的真昼にはなく、2〜4時間ほど先行した形をとる。この放射線耐性リズムと同一位相で走る抗酸化能リズムも振幅は小さいながら認められる。

実験にはユーレナを6〜8万細胞/mLになるまで25\(^\circ\)C、6 klx 恒明（白色蛍光灯）で培養し、恒暗に移す。恒暗中の様々な時点で \(\gamma\)-線や220 MeV 12Cイオン（以下Cイオン）を照射し、中性赤を用いて生死判別を行った。その生存率をもって耐性の基準とした（Cイオンは照射直後、 \(\gamma\)-線は一日後）。

\(\gamma\)-線とCイオン照射を行う温度（0、25\(^\circ\)C）は生存率に影響しない。これはUV照射の場合も同様である。

これに対し、照射後に低温（4\(^\circ\)C）放置した場合には、25\(^\circ\)Cで保存した場合よりも10〜60 %ほど高い生存率を示し、温度依存的な生命活動が死を促進する可能性が示された。このことは、 \(\gamma\)-線照射の場合に頑著に認められる。

\(\gamma\)-線照射は細胞死を含むすべて照射中及び照射にのみ生起するので、この点については比較対照外である。

\(\gamma\)-線耐性増強には還元型グルタチオン、GSH-エチルエステル、L-ヒステジンなどが有効であった。また、\(\beta\)-カロチンやリコピンも少なからず有効であった。

Cイオン耐性増強に有効であったのはステアリン、N-アセチルステアリン、ラチチン、アスコルビン酸、還元型グルタチオン、GSH-エチルエステル、トロロックス、\(\beta\)-カロチンやリコピンなどの抗酸化剤である。したがって、 \(\gamma\)-線耐性やCイオン耐性には水溶性及び脂溶性のラジカル消去能と一重酸素消去能が総合的に関与しているものと考えられる。ただし、両耐性は互いに異なる抗酸化作用も関係している。例えば、 \(\gamma\)-線耐性増強に有効な抗酸化剤はCイオン耐性増強にも有効であったが、逆にCイオン耐性増強に有効なアスコルビン酸、N-アセチルステアリン、ラチチン、トロロックスなどは \(\gamma\)-線耐性増強には殆ど無効である。

また、還元型グルタチオン、ステアリンや \(\beta\)-カロチンを含む抗酸化剤の組み合わせ投与により、600 Gy程度の低線量照射の場合、 \(\gamma\)-線耐性やCイオン耐性も非酵素的抗酸化能だけで説明できるが、高線量照射の場合には、抗酸化能以外の要因も関与している可能性が示唆される。更に、分子量の異なるFITC-デキストランを用い、放射線によるユーグレナの死滅に細胞膜の物理的損傷の有無を調べたところ、 \(\gamma\)-線照射の場合には高線量でもFITC-デキストランを透過させずの損傷は生じないことが明らかになった。これに対し、Cイオン照射の場合には1,500 Gy以上の線量でFITC-デキストランの取り込みが顕著に確認された：10 kDのもので50%のユーグレナに、20 kDで10〜15%のユーグレナに取り込まれる。したがって、Cイオン照射による致死作用には物理的生体膜損傷が関与していると考えられることを示すが、高線量Cイオン耐性には抗酸化能は無縁であるという事実を示している。
3-52 Detection of Damage Using ESR Method to Irradiated Wheat Flour

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Irradiation of food is a non-thermal treatment and a method of cold pasteurization. This pasteurization technique retains the quality of foods as prepared and harvested. Recently, the use of this sterilization technology by irradiation through various electromagnetic waves or rays of elementary particles is gaining wide applications. Since irradiation induces free radicals, consumer believes that irradiated foods can be the threatening of public health.

Electron spin resonance (ESR) spectroscopy has been applied for the evaluation of irradiation on the wide variety of foods. We have reported on a new ESR protocol for the analysis for irradiated foods 1).

In the present study, we will report on the radiation-induced radicals containing in wheat flour using ESR spectroscopy. We shall describe new radical species in the irradiated wheat flour by comparison with non-irradiated wheat flour. Attempts have been made on the irradiation damage of radicals.

The flour sample of wheat was the products of Gumma, Japan. The nutritional contents of the wheat flour consist of starch (70 wt %) and protein (14 wt %). In order to avoid the anisotropic EPR spectral dependence by the microcrystalline of grain in wheat flour, we employed a separation method of powder sizes of less than the 200 meshes. Thereby, the identical ESR signals were detectable irrespective of the sample tube rotation. The specimens were irradiated at the JAEA Takasaki at room temperature (ca. 20 ºC). The dosage level of the gamma ray was controlled by the irradiation period. We selected the three dose levels, i.e., 10, 30 and 50 kGy.

ESR measurements were carried out using an ESR spectrometer (Jeol, JES-FE1XG and JFS-FA200). All the ESR spectra were recorded at the X-band (9.3 GHz). In order to detect full view of all the radical species of wheat flour, we employed two kinds of field sweep, 250 ± 250 mT and 320 ± 100 mT.

The ESR spectrum of wheat flour before irradiation consists of a sextet centered at g = 2.0 and a singlet signal at the same g-value position (Fig.1). The first one is attributable to a signal with hyperfine (hf) interactions of Mn$^{2+}$ ion (hf constant: 7.4 mT). The second is originated from carbon-centered radical.

Upon gamma ray irradiation, however, a new signal with two triplet lines at the low and high field ends was detected in wheat flour on top of the Mn$^{2+}$ sextet lines (Fig.2). We analyzed the triplet ESR lines as powder spectra (rhombic g-tensor symmetry) with nitrogen (14N) hyperfine interactions. This indicates that a new organic radical was induced in the conjugated protein portion of wheat flour by the gamma ray irradiation. Intensity of the organic free radical at g = 2.0 detected in irradiated wheat flour increased with the degree of irradiation.

Reference
Preparation of Acellular Arteries by Gamma Irradiation

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Synthetic materials, such as ePTFE or Dacron are not suitable for reconstructing the small-diameter arteries. In contrast, tissue engineering (TE) graft has been attracting great attention recently. One of the promising approaches of TE grafts is using acellular tissues. The acellular scaffold may have the same structure and characteristics as the natural tissue and can be replaced with the host tissues. In this study, we prepared the acellular scaffold using gamma irradiation, which is expected to induce the apoptosis of the cells.

Preparation of Acellular Arteries by Gamma Irradiation

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Synthetic materials, such as ePTFE or Dacron are not suitable for reconstructing the small-diameter arteries. In contrast, tissue engineering (TE) graft has been attracting great attention recently. One of the promising approaches of TE grafts is using acellular tissues. The acellular scaffold may have the same structure and characteristics as the natural tissue and can be replaced with the host tissues. In this study, we prepared the acellular scaffold using gamma irradiation, which is expected to induce the apoptosis of the cells.

現在、我が国において人工心臓弁は年間1万個、人工血管は5万本が使用されている。屍体から提供されるヒト心臓弁や血管も組織バンクを通じて臨床使用されているが、年間数十件程度に過ぎない。米国では商業利用によって、年間数千件のヒト組織が使用されている。人工素材から作製される移植用組織は生体にとっては異物であり、自己組織と置き換わることは少ない。また、小児患者において血管の成長性が欠如しているという欠点もある。近年、移植後の拒絶反応を避けるために、自己組織と置換される素材を用いた移植血管や、ドイツ・フンボルト大学グループによるプタ脱細胞化組織を用いた再生型人工血管が報告されている。我々は、放射線照射によって細胞を除去した生体組織をバイオスキャホールド（細胞足場材料）として用いた再生型移植用組織の開発を行っている。

移植組織片は以下のようにして作製した。生後4ヶ月、体重約10 kgのクラウン系ミニプタ（㈱ジャパンファーム、鹿児島）から清潔下にて下行大動脈を採取した。PBSによる洗浄後、PBSを満たした滅菌容器に封入し、10, 30, 100, 300, あるいは1000 Gyのガンマ線を高崎量子応用研究所のCo-60照射施設を用いて照射した。吸収線量率は、それぞれ100, 300, 1000 Gy/hである。照射後、PBSをベースとする洗浄液にて2週間洗浄した。洗浄液は、適宜交換した。Wistarラット（7週令）の皮下部位に上記脱細胞化ミニプタ大動脈を埋入し、2週間後に取り出し、ヘマトキシリン-エオジン染色、マクロファージに対するCD68免疫染色にて組織学的検討を行った。種々のガンマ線量による組織脱細胞化の基礎的検討を残存DNA定量試験、力学試験にて行った。残存DNA定量試験では、100あるいは300 Gy以上の照射では大幅なDNA減少傾向が見られた。また、力学特性は、破断强度並びに弾性率とも大きな影響は見られないかった。すなわち、300 Gy以上のガンマ線照射により、洗浄処理することによって、循環器系組織内の細胞はほぼ完全に除去できると思われた。

次に、ガンマ線照射によって作製した脱細胞化ミニプタ大動脈の有効性を、ラット皮下への埋入試験によ
Radionuclide-based imaging techniques enable us to obtain images of the kinetics of the transport of water and nutrients in plants, and hence determine plant physiology. In particular, by using the positron-emitting tracer imaging system (PETIS), which was designed for studying plant physiology and agriculture, many experiments have been performed for the imaging of the distribution and translocation of the nutrients using positron emitting tracers. The kinetics of $^{11}$C make it a potential candidate for estimating the physiological function parameters of photosynthesis. By using $^{11}$CO$_2$, the kinetics of photoassimilation and of the flow of sucrose, which is synthesized as a photosynthate, through sieve tubes have been obtained as movies comprising serial images. We have analyzed the exchange of carbon compounds inside and outside a test leaf, based on the $^{11}$C-labeled tracer kinetics in the compartmental model. An analytical solution of the carbon tracer balance was obtained by the following equation; as follow:

\[ L(t) = K_{1} \cdot C_{i}(t) \otimes e^{-(k_{3}+\lambda)t} \]  

(Eq. 1)

where, $L(t)$ is the leaf response time-activity curve, which indicates the time variation of $^{11}$C concentration in leaf; $C_{i}(t)$, the input time-activity curve; $\lambda$, the physical decay of $^{11}$C; and $\otimes$, the convolution integral. Further, photoassimilation ($K_{1}$) and the sucrose export ($k_{3}$) rate constant were estimated by kinetic analysis$^{13}$.

In order to produce parametric images of photosynthetic functions from the PETIS movie data, we performed a pixel-by-pixel analysis by the present compartmental method. To construct the images depicting photoassimilation and the sucrose-export images from the estimated values of the parameters $K_{1}$ and $k_{3}$, approximately 10,000 pairs of time-activity curves were fitted to the Eq. 1 by the nonlinear least squares method. By this analytical method, 2 bioactive distribution images were produced from a single movie data.

Shown in Fig. 1 are the parametric images of 2 photosynthetic processes namely, photo-assimilation and sucrose export. The $K_{1}$ image indicates that photoassimilation was distributed uniformly over the entire leaf. In contrast, the $k_{3}$ image indicates that sucrose export was not distributed uniformly. The sucrose export ability at the tip was greater than that at the base. This seems attributed to the well-known fact that the base of the leaf is the region of growth. It implies that sucrose produced at the base of the leaf was probably consumed there itself for the growth, and that produced at the tip was mainly transported to the other parts of the plant.

In summary, we have performed imaging using PETIS and kinetic analysis for studying the processes of plant physiology, such as photosynthesis in living systems mainly in response to the environment, and processes involved in the anisotropic maturation of leaf tissues$^{21}$.

References

Fig. 1 Parametric images of photosynthetic functions. The left image on the left indicates the distribution of photo assimilation ($K_{1}$) and the image on the right indicates the distribution of sucrose export in different regions of the leaf ($k_{3}$).
3-55 Non-invasive Estimation of Phloem Flow Velocity under Cadmium Stress Using the Positron Emitting Tracer Imaging System

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Cadmium (Cd) is a major environmental pollutant, which accumulates in agricultural soils, and is a toxic element to all organisms. In plant, the most sensitive symptom of Cd toxicity is stomata closure, followed by the inhibition of photosynthesis. However, it has been reported that Cd treatment increases carbohydrate content of source organ. This contradictory result could be due to the inhibition of translocation of photoassimilates by Cd. However, direct evidence has not been reported.

In this study, we estimated a phloem flow under Cd stress using the positron emitting tracer imaging system (PETIS), quantitatively and non-invasively. In an experiment, 11CO2 was supplied to leaves of rice plants and the translocations of 11C-photoassimilates were monitored.

Rice plants (Oryza sativa L. cv Nipponbare) were germinated and grown hydroponically in nutrient solution (without Cd) for 4 weeks. Plants were transferred to the nutrient solution containing 0 (control), 1, 10 and 100 µM of Cd, and cultured for 1 day before PETIS experiment.

Five to six of Cd-treated plants were placed between a set of PETIS detectors in a growth chamber. The upper half of the largest expanded leaf of each plants was inserted into a “gas-cell”, a clear acrylic box for feeding 11CO2 (100 MBq) gas (Fig.1). Time course of 11C-radioactivity (Time-Activity Curve) in the basal region was generated from the serial images of each rice plant (Fig.1). The value of the x-axis intercept of approximated line was defined as 11C-arrival time to basal region.

We performed 6 sets of PETIS experiments using 5 ~ 6 different rice plants treated with 0, 1, 10, 100 µM of Cd. The arrival times of 11C-photoassimilates to the basal region of each plant were analyzed (Fig. 2). Phloem flow velocities were also estimated using the 11C-arrival time and the distance from the bottom of gas-cell to the basal region.

Arrival time of 100 µM Cd-treated plants was significantly different from that of control and 10 µM Cd-treated plants (p < 0.05). This result indicates that Cd exposure for 1 day causes the decrease of phloem flow velocity in rice plant. Now we are also examining the rate of photoassimilation in source leaf and the concentration of photoassimilates in phloem sap under Cd stress. These data will provide the basis of Cd toxicity to plant.

This study indicates the utility of estimation of phloem flow velocity using PETIS and simple data analysis. This method can be applied for studying influences on phloem transport against other environmental conditions.

Fig.1 Time-Activity Curve for accumulation of 11C-photoassimilates in the basal region of rice plants treated with 0, 1, 10, 100 µM of Cd. The figure on the left shows the image of 11C-radioactivity captured by PETIS and the picture of tested plants.

Fig.2 Effect of Cd stress on 11C-arrival time to basal region. Means with the same letter are not significantly different (p < 0.05). n=7-8
Nitric acid and ammonium absorbed by the roots, and carbon dioxide fixed on the leaves, are metabolized, distributed and accumulated into the plant organs. Correlation between carbon assimilation and nitrogen assimilation is one of the important factors regulating plant growth.\(^1, 2\) The Positron-Emitting Tracer Imaging System (PETIS) and tracers of major nutrition have been an available system to obtain movies of tracer in short-term distribution non-invasively. Therefore, we tried to visualize the movement of carbon and nitrogen nutrition in a whole plant.

Soybean (\textit{Glycine max} [L.] Merr. cv. Enrei) plant inoculated with rhizobium had been hydroponically cultured with MS medium for 25 days, and was used for the PETIS experiment (Fig.1). Three tracers, \(^{11}\text{CO}_2\), \(^{15}\text{NO}_3^-\), and \(^{15}\text{NH}_4^+\) were produced by bombarding targets (\(^{11}\text{CO}_2\): nitrogen gas, \(^{15}\text{NO}_3^-\): pure water, \(^{15}\text{NH}_4^+\): pure water with a small amount of ethanol) with proton beam delivered from the TIARA AVF cyclotron. Soon after approximately 30 MBq of \(^{11}\text{CO}_2\) was fed to the leaves, PETIS images were acquired for 2 hours. After first imaging, 100 MBq of \(^{15}\text{NO}_3^-\) was fed to the roots and PETIS images were acquired for 90 min. Subsequently, 16 MBq of \(^{15}\text{NH}_4^+\) was fed to the roots and images were acquired for 90 min. The interval of each imaging was about 30 min to allow radioactivity to decay at the background level.

As a result, a movie of distribution of \(^{11}\text{C}\)-labeled photoassimilates in the plant organs and nodules, and movies of translocations with \(^{15}\text{NO}_3^-\) and \(^{15}\text{NH}_4^+\) were acquired (Fig.2). In other words, the real time imaging of translocation of \(^{11}\text{C}\) labeled carbon source and \(^{15}\text{N}\) labeled nitrogen in an intact nodulated soybean plant was demonstrated.

Furthermore, nitrogen fixed by nodule was important factor regulating plant growth in soybean plant. An imaging experiment on N\(_2\) fixation of nodule and fixed nitrogen translocation in soybean is currently in progress by using PETIS and \(^{15}\text{N}_2\) gas.

References
\(^1\) H. Fujikake et al. J Exp Bot. 2003 May; 54 (386) 1379
\(^2\) S. Ito, N. Ohtake, K. Sueyoshi and T. Ohyama
Cd is a harmful heavy metal element. It causes various obstacles in the human body when it is taken in via the food. Cd is also famous as a causative substance of the itai-itai disease. Recently, there is the international movement which makes the reference level of cadmium content in the food harsh. It has become national urgent business to establish the agricultural technology which decreases the cadmium content in the farm products.

In order to decrease the cadmium content in the farm products, two technologies are focused recently. One is called “phytoremediation” which cleans up the cultivation soil using absorption by special plants. The other is to modify the crop plants not to accumulate cadmium in their edible parts. However, these technologies are not practical yet. For realization of these technologies, it is necessary to elucidate the mechanism of transport of Cd in the plant body.

In this research, we visualized Cd transport in the plant body by using Positron Emitting Tracer Imaging System (PETIS). We tried to elucidate the mechanism of Cd transport in the whole plants by analyzing the experimental data which is obtained by PETIS.

$^{107}$Cd which is positron-emitting nuclide was used for PETIS experiment. Production method of $^{107}$Cd was established by Ishioka et al. Sorghum plants ($Sorghum vulgare$ L.) and oilseed rape plants ($Brassica napus$ L.) were grown hydroponically for two weeks after sowing. PETIS experiment was performed inside the growth chamber where we can control growth conditions of plants completely.

After setting plants inside the chamber, PETIS experiment was started by adding purified $^{107}$Cd in hydroponic solutions. Time-series images of the $^{107}$Cd distribution in the aerial part and upper part of the roots of plants were obtained with the PETIS apparatus. Time courses of the absorption of water and the tracer ($^{107}$Cd) by plants were also monitored. Time-activity curves, temporal changes of the tracer amount in specific regions of interest (leaves, stems, and so on) were generated from PETIS data. These data were combined and analyzed.

We succeeded to obtain fine serial images of Cd transport in sorghum plants (Fig. 1) and oilseed rape plants (Fig. 2). We could observe that $^{107}$Cd signals arrive in the aerial parts of both plants in a few hours after the start of the experiment. Patterns of accumulation of $^{107}$Cd signals in sorghum plants were similar to those in rice plants. Strong signals of $^{107}$Cd were seen in the basal region of the shoot, which consists of a short stem, nodes and meristems. Interestingly, $^{107}$Cd also accumulated in the node strongly in oilseed rape plants although these two plants have different structure as a monocot and a dicot. The analysis of time-activity curves revealed that pattern of accumulation of $^{107}$Cd changes in development in the oilseed rape plants.

References
 Imaging of Translocation and Spatial Distribution of the Assimilated Carbon and Nitrogen in the Host–root Parasite System by PETIS

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Translocation and distribution of $^{13}$C, $^{13}$N-nitrate or $^{13}$N-ammonium in the red clover (Trifolium pratense L.) parasitized by small broomrape (Orobanche minor Sm.) was examined by PETIS. The PETIS images acquired periodically after $^{13}$CO$_2$ exposure indicate that Orobanche is a big ‘sink’ of the assimilated $^{13}$CO$_2$ by host plant. Accumulation of $^{13}$N applied as $^{13}$NH$_4^+$ in Orobanche tubercles was higher than that applied as $^{13}$NO$_3^-$, indicating that $^{13}$NH$_4^+$ stay longer in Orobanche tubercles than $^{13}$NO$_3^-$. The differences in the accumulation may be due to the rapid metabolism of ammonium to amino acids.

Sharing photoassimilates and absorbed nitrogen-compounds with host plants is of primary importance for the survival of root parasites. Direct imaging of translocation and spatial distribution of the assimilated carbon and nitrogen in the host–parasite system is the most convincing approach for the study of host–parasite interactions. However, each host-parasite system is unique and may be different from others, and therefore, it is quite difficult to obtain reproducible results. Consequently, a noninvasive and repeatable imaging system using the radioactive nuclides having short half-lives is needed. The positron-emitting tracer imaging system (PETIS), designed for the use in plant science to study the translocation and distribution of nutrients by using radioisotope tracers ($^{14}$C, $^{15}$N, etc), allows us to perform noninvasive, repeatable, and quantitative analyses of nutritional sharing between host plants and root parasites. In the present study, translocation and distribution of $^{13}$CO$_2$, $^{13}$N-nitrate or $^{13}$N-ammonium in the red clover (Trifolium pratense L.) parasitized by small broomrape (Orobanche minor Sm.) was examined by PETIS.

The PETIS images acquired periodically after $^{13}$CO$_2$ exposure indicate that Orobanche is a big ‘sink’ of the assimilated $^{13}$CO$_2$ by host plant (Fig.1). Based on the image analysis of the $^{13}$C radioactivity in the host–parasite system, 15% of the carbon assimilated by host plant accumulated in Orobanche 30 to 40 min after the $^{13}$CO$_2$ exposure. In addition, it seemed that Orobanche tubercles absorbed $^{13}$NH$_4^+$ more strongly than $^{13}$NO$_3^-$, indicating that $^{13}$NH$_4^+$ stay longer in Orobanche tubercles than $^{13}$NO$_3^-$ (Fig.2). However, relative ratios of radioactivity in Orobanche tubercles to that in the root area were very similar in both the $^{13}$NO$_3^-$- and $^{13}$NH$_4^+$-fed host–parasite systems, suggesting that Orobanche tubercles absorb $^{13}$NH$_4^+$ and $^{13}$NO$_3^-$ with similar affinities. The differences in the accumulation may be due to the rapid metabolism of ammonium to amino acids.
3-59 Fe and Zn Translocation from the Roots to the Shoots of Rice Monitored by Positron Emitting Tracer Imaging System (PETIS)

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1. Introduction
Iron (Fe) and zinc (Zn) are essential nutrients for plants. However, little is known how Fe and Zn are translocated in rice plants especially to the ear after absorption from roots. In addition, the real-time measurement of translocation of Fe and Zn has not been documented in plants. To study the pathway of Fe and Zn to the ear, we traced $^{52}$Fe and $^{62}$Zn in rice plants (Oryza sativa L. cv. Nipponbare) of 3 weeks after flowering by positron emitting tracer imaging system (PETIS).

2. Experimental procedure
Rice (Oryza sativa L. cv. Nipponbare) were hydroponically cultured in a growth chamber under a mixture of incandescent and fluorescent lamps with a 14 h light (30 °C)/10 h dark (25 °C) regime and a photon flux density of 320 $\mu$mol m$^{-2}$ s$^{-1}$. The nutrient solution was renewed once a week. The absorption experiments were performed about 3 months after germination and 3 weeks after flowering. A rice plant was supplied with 20 mL of culture solution in polyethylene bags. The plants and the bags were fixed an acrylic board and placed between a pair of PETIS detectors in a chamber at 30 °C and 65% humidity and a light density of 320 $\mu$mol m$^{-2}$ s$^{-1}$. $^{52}$Fe$^{3+}$-deoxymugineic acid (DMA) (about 2 MBq) or $^{62}$Zn$^{2+}$ (about 20 MBq) were added to culture solution. The shoots were monitored by PETIS for 14 h.

3. Results and discussion
In order to clarify the pathway of Fe and Zn from the roots to the ear of rice, $^{52}$Fe$^{3+}$ or $^{62}$Zn was translocated to the shoots within 14 h (Fig. 1, Fig. 2). In particular, the nodes were strongly labeled. Both $^{52}$Fe and $^{62}$Zn were preferentially accumulated in the lower node until 2 h and 5 h after absorption, respectively. However, the radioactivity of upper node was higher than that of lower node in the last results. It was reported that the nodes contained the elliptical, diffuse and anastomosing vascular bundles, and the xylem transfer cells were found in the elliptical bundles. Therefore, $^{52}$Fe and $^{62}$Zn might be transferred from xylem to phloem in the nodes.

Reference
1) S. Y. Zee et al., Aust. J. Bot. 20 (1972) 41.
Impact of Aluminum on the Photosynthesis and Translocation of Photoassimilates in Rice Seedling. 
-Imaging-based Analysis by PETIS-

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Accumulation, translocation and distribution of \(^{11}\text{C}\)-labeled photoassimilates in the rice (\textit{Oriza sativa}, cv. Nipponbare) seedling treated with or without aluminum (Al) was examined by PETIS. The PETIS images acquired periodically after \(^{11}\text{CO}_2\) exposure indicate that the long-term exposure promote the accumulation of \(^{11}\text{C}\)-labeled photoassimilates and its translocation toward roots. In contrast, the short-term exposure attenuate both. Similar results were observed in the seedlings of tobacco, suggest that Al-dependent modulation on the rate of photosynthesis and the following translocation of \(^{11}\text{C}\)-labeled photoassimilates is the common feature of land plants.

Thirty to 40% of arable land in the world has been estimated to be acidic soils. In addition global industrialization has triggered huge exhaustion of acidic compounds into the atmosphere, which expanded the area of acidic soils in all over the world. In the regions where acidic soils are prominent, most of the crops or pastures are disturbed their growth and development. Aluminum (Al), the most abundant metal element in soils, is solubilized as free ions under acidic conditions and exhibits toxicity to plants. A variety of plants can grow in acidic soils by secretion of organic acids from root apices, which excludes and/or detoxifies Al ion in the apoplast. To support the efflux of organic acid from root apices under Al stress, sugars should be transported to the root apices, but how the rate and amount of sugar-translocation are regulated is still obscure.

To understand that, \(^{11}\text{CO}_2\) was applied to the entire seedlings which settled in a air-tight clear-acrylic box (inside dimensions: 8 cm in length, 5 cm in width, 1 cm in depth), and the box was positioned in the middle of the PETIS detectors. In rice seedlings, a major crop of monocots, were pre-treated with Al, \(^{11}\text{CO}_2\) fixation and sugar translocation toward roots were enhanced by the long-term exposure (31.5 h) to AlCl\(_3\) (100 µM) in acidic condition. In contrast, relatively short-term exposure (25.5 h) attenuate those and those were recovered in transition state (28.5 h), as observed in tobacco seedlings (reported in the previous report)(Fig.1). These results suggest that the stimulation of photosynthesis in leaves by Al exposure to roots and subsequent translocation of photoassimilates toward root part is a common feature of both dicots and monocots.

Fig.1 Aluminum-dependent modulation of photoassimilates-distribution in rice seedlings. Hydropotonically-grown rice seedlings (20-day old) were treated without (control, right two in each panel) or with 100 µM AlCl\(_3\) (+Al, left two in each panel) in modified Ruakura medium (pH 4.3) for the indicated durations. Then the entire seedlings were expose to \(^{11}\text{CO}_2\), and the distribution of \(^{11}\text{C}\)-labeled photoassimilates was monitored for 120 min. The radioactive counts integrated for every 12 min from a start till 120 min are shown.
3-61 Cyclotron Production of PET Radionuclide: No-carrier-added Bromine-76 with Protons on Natural SeO$_2$ and KBr Targets

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In recent years Positron Emission Tomography (PET) has become established as particularly attractive tracer for cancer detection using positron-emitter. The increasing use of PET for clinical diagnosis, drug development and biological research has prompted many chemists to develop new labeling and purification methods. The PET radionuclides such as $^{11}$C, $^{15}$N, $^{18}$O and $^{18}$F have been used for imaging agent, which are short half-lives; $^{18}$F with the longest half-life is 110 minutes. On the other hand, the positron-emitter with a longer half-life that can be applied to the PET diagnosis based on a nuclear characteristic is proposed besides above-mentioned nuclides. $^{76}$Br is of potential interest in nuclear medicine: $^{76}$Br has been suggested as a halogen label with a convenient half-life ($T_{1/2} = 16.1$ h) and a sufficient $\beta$-branching ratio (57.2 %) in PET. Here we present the development of a direct and an indirect production methods for $^{76}$Br.

$^{76}$Br was produced from the direct and indirect nuclear reactions ($^{76}$Se(p,n)$^{76}$Br, $^{79}$Br(p,4n)$^{76}$Kr ($T_{1/2} = 4.6$ h) $\rightarrow^{76}$Br). In the direct method, 6 ml of SeO$_2$ solution with the natural isotope composition was irradiated for 20-30 minutes with proton beams (20 MeV) of 1 $\mu$A. The generated $^{76}$Br was separated from the irradiated solution by anion exchange method. In the indirect technique, 1.2 g of NaBr pellet (10 mmol) with the natural isotope composition was irradiated for 5-10 minutes with proton beams (65 MeV) of 0.5-1 $\mu$A. The irradiated pellet was dissolved in 10 ml of H$_2$O. After dissolving the pellet 5 ml of 10% H$_2$SO$_4$ was added in the solution. The resulting $^{76}$Kr was transported to the upper vial with H$_2$ carrier gas and trapped in the 5 ml of H$_2$O (Fig. 1). After the decay of $^{76}$Kr the generated $^{76}$Br was collected. The $^{76}$Br produced by both methods was measured with the high-pure Ge detector in each solution.

The production of $^{76}$Br with proton induced reactions on natural SeO$_2$ and NaBr targets can be accomplished. These $^{76}$Br yields are summarized in Table 1. The production yields were found to be 21-24 kBq/$\mu$A$\cdot$min with SeO$_2$ target and 126 MBq/$\mu$A$\cdot$min with NaBr target (EOB). In addition, the maximum production yield was calculated using the value of maximum beam current and irradiation time at TIARA AVF cyclotron. It was estimated that the amount of $^{76}$Br was about 240 MBq in the use of SeO$_2$ target, and was about 360 MBq in the use of NaBr. In the comparison of production yields, an indirect method is more advantageous.

In a clinical application of $^{76}$Br, it is necessary to be sufficient radioactivity for subsequent labeling. Further research into the development of the production method using enriched $^{76}$Se target is currently underway.

![Fig. 1 Separation of Kr from NaBr.](image)

Table 1
Recovery yields of $^{76}$Br with SeO$_2$ and NaBr targets and maximum activity using TIARA AVF cyclotron

<table>
<thead>
<tr>
<th>Nucler reaction</th>
<th>Beam energy</th>
<th>Target</th>
<th>$^{76}$Br yield (kBq/$\mu$A$\cdot$min)</th>
<th>Irradiation condition</th>
<th>$^{76}$Br Activity (calculated)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{76}$Se(p,n)$^{76}$Br</td>
<td>20 MeV</td>
<td>0.1% SeO$_2$</td>
<td>21</td>
<td>5 $\mu$A $\times$ 16 h</td>
<td>120 MBq</td>
</tr>
<tr>
<td>(direct method)</td>
<td></td>
<td>10% SeO$_2$</td>
<td>24</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{76}$Br(p,4n)$^{76}$Kr</td>
<td>65 MeV</td>
<td>NaBr</td>
<td>126</td>
<td>3 $\mu$A $\times$ 16 h</td>
<td>360 MBq</td>
</tr>
<tr>
<td>(indirect method)</td>
<td></td>
<td></td>
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</table>
PET is superior in quantitative measurement, so 18F-FDG-PET is most valuable tool for tumor diagnosis. Although several positron emitters have been used for PET, their uses are limited for their short half-lives. Compared with these radionuclides, 64Cu and 76Br have appropriate properties (76Br: T1/2 = 16.1 hr, 64Cu: T1/2 = 12.7 hr) and they may have great potentials for PET utility. In this study, we synthesized monoclonal antibody (mAb) labeled with 64Cu or 76Br and evaluated their potential for tumor diagnosis with PET. The results of tumor localization studies show that 64Cu or 76Br labeled mAb were highly accumulated to tumor. From these data, the use of 64Cu and 76Br has great advantage for PET utility.

PET (ポジトロン断層画像撮像法)はSPECT（シングルプラット断層画像撮像法）、シンチカメラに比べて定量性、解像度の点で優れており、有用な画像診断法として注目されてきた。しかしながらPETに用いられる放射性核種（RI）はそれも半減期が短く（18F：110分、15O：2分、18N：10分、13C：20分）、その利用にはサイクロトロンの設置が必要になるなど費用がかかり、普及が制限されていた。近年、がん診断に注目されてきたが、PET用放射性薬剤の開発、製薬企業による供給が可能ながん診断薬、脳機能診断などに有効な放射性薬剤の開発に適したリジオノマー（ RI）が開発された。この中でもPET用として発現の半減期（現在臨床で用いられている18Fの半減期は約3時間）を有する放射性核種（RI）の開発に適したと考えられる。

一方PET診断用RIとして半減期：12.7時間の64Cuおよび半減期：16.1時間の76Brの開発が計画された。64Cuは金属核であることから化合物への導入にキレート結合が必要とするが、比較的安定な放射性薬剤を得ることが可能である。64Cuの製造は18N(p,n)64Cu反応により行うが、64Cuをモノクローナル抗体（NuB2）に標識し、マウスを用いてそのがん診断薬としての有用性を評価した。抗体とRIの組合せには1,4,8,11-tetraazacyclotetradecan-1,4,8,11-tetraacetic acid (TETA)を用いた。

一方76Brは18Fや121Iと同じハロゲンであり、これまでに開発されたがん診断、脳機能診断などに有効な18Fあるいは121I標識放射性薬剤をベースとした放射性薬剤の開発に適したRIであると考えられる。76Brの製造は76Se(p,n)76Brあるいは7Br(p,4n)7Kr→76Krにより行った。分離精製後、76BrをNuB2に直接標識し、64Cuと同様の方法により動物体内動態を評価した。

64Cu-TETA-NuB2の標識率は約90%、精製後の放射化学的純度は99%以上であった。がんを移植したマウスに64Cu-TETA-NuB2（約1µCi匹）を投与したところ、腫瘍への高いRI集積を認めた2)（投与2日後で約13%dose/g、Fig.1A）。一方76Br-NuB2の標識率は約10%と低く、精製後の放射化学的純度は約95%であった。64Cu同様、担がんマウスに76Br-NuB2（1～4μCi匹）を投与し、その体内動態を調べた結果、76Br-NuB2も腫瘍へ高く移行することを認めた3)(投与2日後で約10%dose/g、Fig.1B)。

以上の結果より、新規PET用RIを用いたがん診断薬の有用性が示された。

References
Incident Energy Effect of the Production Yield of Endohedral $^{133}$Xe-Fullerene by Ion Implantation

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Endohedral fullerenes encapsulating a radioisotope within a fullerene cage have a possibility of being new radiopharmaceuticals. We have prepared endohedral $^{133}$Xe-fullerenes by ion implantation$^1$. Xenon-133 ($T_{1/2} = 5.243$ d) would be useful for cancer therapy because it emits 0.346-MeV $\beta$ rays (maximum range in water: 1.0 mm) followed by 81-keV $\gamma$ rays and conversion electrons associated with the $\gamma$ transition. However, we could not obtain efficient production of endohedral $^{133}$Xe-fullerene. To achieve more efficient production, conditions of the ion implantation, especially incident energy, has to be studied. In the present paper, we describe incident energy effect on the production yield of endohedral $^{133}$Xe-fullerene.

Fullerene targets for ion implantation were made by vacuum evaporation of $C_{60}$ on Ni foils. Implantation of $^{133}$Xe$^+$ ions into the targets was carried out with an isotope separator at acceleration energies of 30 keV. For retardation of incident energy, the high voltage of 15kV was supplied from a retardation power supply to the fullerene targets: incident energy became 15 keV. Doses were in the rage of $1\times10^7$ to $1\times10^8$ cm$^{-2}$. After ion implantation, the fullerene part on the target was dissolved in o-dichlorobenzene. The solution was filtered through a membrane filter to remove insoluble materials. The filtrate was analyzed by high performance liquid chromatography (HPLC) with a Cosmosil Buckyprep column. Elution was done with o-dichlorobenzene at a flow rate of 1 ml/min. The concentration of fullerene in the effluent was continuously monitored by a UV detector. The effluent after UV measurement was collected in glass vials each for 1 min until 20 min and the $^{133}$Xe radioactivity in each vial was measured by $\gamma$-ray spectrometry.

Figure 1 shows elution curves of $^{133}$Xe and $C_{60}$ obtained from the sample bombarded with $^{133}$Xe ions of 15 keV at a dose of $2.0\times10^{12}$ cm$^{-2}$. The strong correlation observed between the $C_{60}$ and $^{133}$Xe peaks corroborates the formation of $^{133}$Xe@$C_{60}$. HPLC data obtained from the other samples also corroborate the formation of $^{133}$Xe@$C_{60}$. In order to evaluate the efficiency of the formation of $^{133}$Xe@$C_{60}$, the yield of $^{133}$Xe@$C_{60}$ was defined as a percentage of the radioactivity of $^{133}$Xe@$C_{60}$ to that of $^{133}$Xe implanted into the target. The radioactivity of $^{133}$Xe@$C_{60}$ was obtained from the sum of the $^{133}$Xe radioactivities in the fractions from 3 to 10 min. The yields of $^{133}$Xe@$C_{60}$, each of which is derived from one experiment, were plotted as a function of $^{133}$Xe dose in Fig. 2. The solid and broken straight lines in those figures, which were fitted by the method of least-squares on log-log scales, are $y=(10.18\pm0.09)-(0.842\pm0.007)x$ and $y=(9.6\pm0.8)-(0.80\pm0.06)x$ on 15 and 30 keV, respectively. For both dose studied, the yields of $^{133}$Xe@$C_{60}$ were found to decrease with increasing dose. Because an increase of the dose leads to an increase of the number of damaged fullerene molecules as reported by Kastner et al.$^2$ from their Raman spectra measurements of fullerene targets bombarded with energetic-ions, those dose dependences of the production yields can be ascribed to amorphization of $^{133}$Xe@$C_{60}$.

From comparison between both lines one can note that the yield of 15 keV is about 25% larger than that of 30 keV for the same dose in the rage of $1\times10^{12}$ to $1\times10^{14}$ cm$^{-2}$. Those energy dependences of the production yields can be also ascribed to the amorphization of $^{133}$Xe@$C_{60}$. The yields of the $^{133}$Xe@$C_{60}$ should be more increased by decreasing incident energy.

References

Analysis of Elements in Tumors and Normal Tissues following Irradiation by Micro-PIXE

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The distribution of elements at the cellular level can be imaged by an in-air micro-particle-induced X-ray emission (micro-PIXE), but its application at the tissue level has been very limited. We compared the distribution of elements in tumors and normal tissues examined by the micro-PIXE with light-microscopic findings \textsuperscript{1)}. Furthermore, we analyzed the changes in the distribution of elements caused by cell death before and after irradiation.

Human ependymoblastoma, glioblastoma, and small cell lung cancer (SCLC) subcutaneously transplanted into male nude mice and normal organs (spleens, livers, kidneys, and testes) of mice were examined. X-ray irradiation (10 or 20 Gy) was administered, and the tumors or normal tissues were excised 3, 4.5, 6, 24, and 72 hours after irradiation. Frozen sections produced with a thickness of 5 and 10 micrometers were fixed by rapid drying and examined using an in-air micro-PIXE. After the examination, the samples were stained by hematoxylin-eosin, and observed by light microscopy to compare the distribution of elements and histological findings.

In the control (non-irradiation) groups, phosphorus (P), potassium (K), chlorine (Cl), and sulfur (S) were observed in the regions, corresponding to viable cells of the 3 tumors and normal tissues, and these elements seemed to be distributed in the nuclei and cytoplasm. In homogeneous areas of cells, these elements were homogeneously distributed, agreeing well with the morphological findings by light microscopy. But, the resolution of light microscopy was clearly higher than that of the micro-PIXE method. There were no significant correlations between the distribution of calcium (Ca), silicon (Si), or iron (Fe) and morphological findings. In the control groups of glioblastoma and SCLC, small necrotic areas were observed, and P, K, Cl, and S were significantly decreased in these lesions.

In the irradiation groups, a high incidence of TUNEL-positive apoptosis was detected in the ependymoblastoma and spleen 3-6 hours after 10 or 20 Gy irradiation by light microscopy. The incidence peaked 6 hours after, and decreased 24 hours after irradiation. In all ependymoblastoma and spleen samples with a thickness of 5, 10, or 20 micrometers, no significant differences between the irradiation and control groups were detected by the micro-PIXE, and the findings were almost the same as those in the control group or slightly inhomogenous in some samples (Fig.1). In the irradiation groups of glioblastoma and SCLC, small necrotic areas were detected at all excision times, and marked decreases in P, K, Cl, and S were observed in the regions, corresponding to the necrosed lesions and tissue defects. But, there was no significant difference in this change observed in the necrosed lesions between the irradiation and non-irradiation groups. No significant changes of elements were observed in the kidneys, livers and testes by the micro-PIXE, though a few apoptotic spermatogonia were found in the testes. There were no significant changes in the distribution of Ca, Si, and Fe in all tumors and normal tissues.

The light-microscopic morphology of the 3 different types of tumors and 4 organs examined in this study markedly differed, but examination by the micro-PIXE demonstrated almost no differences among the tumors and organs if they do not have necrotic areas. The resolution of light microscopy was clearly higher than that of the micro-PIXE.

In this study, we evaluated the applicability of an in-air micro-PIXE method to analyze of the distribution of elements at the tumor and normal tissue levels, and found that necrotic lesions could be imaged, suggesting the usefulness of this method in biological and medical fields. However, since the resolution of this method is much lower than that of light microscopy and apoptotic cell death was not detected, its improvement is required for evaluating the involvement of elements in microscopic changes.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{spleen.png}
\caption{Spleen; 6 hours after 20Gy irradiation.}
\end{figure}

\textbf{Reference}

Investigation of Cisplatin Sensitivity in Esophageal Squamous Cancer Cell Lines and the Localization of Pt Using In-air Micro-PIXE

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The purpose of this study was to elucidate the involvement of CDDP(cis-diammine dichloroplatinum) in ESCC(esophageal squamous cell carcinoma) using in-air micro-PIXE (Particle Induced X-ray Emission). Two human ESCC cell lines–TE-2 and TE-13–were examined for their response to CDDP. TE-2 cells were more highly sensitive to CDDP than TE-13 cells in MTT assay. Dynamic state of element and intracellular metabolism were measured in CDDP treated cancer cells using air PIXE. Using air PIXE, we observed distribution of P and K were localized at cytoplasm, and Br was localized at nuclear site. Air PIXE may be one of the new methods to measure CDDP combined with existing methods. Intracellular concentration of Pt was measured using air PIXE, unfortunately we cannot confirm the difference of distribution of intranuclear Pt in two cells at this moment. Air PIXE may have role in application to predict CDDP sensitivity in cancer cells. Our results using air PIXE has limitation, since it only can be presented by the imaging semi quantitative data. Future study will be needed to establish a new method for precise quantitative data.

References
Direct Visualization and Quantification of Cis-platinum in a Human Lung Cancer Cell Using Micro-PIXE

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We established an imaging method of intracellular CDDP, and also established the method of quantification of intracellular CDDP in a human lung cancer cell line using air micro-PIXE. For the purpose of nuclear labeling, the cells were incubated with $10\mu$M BrdU. After the pulse labeling of BrdU, the cells were exposed to CDDP at a concentration of $1\text{mM}$ for 30 min to 24 hr. Analyses were performed with an in-air micro-PIXE system at the division of Takasaki ion accelerator for advanced radiation (TIARA). For 24 hr exposure of CDDP, there was unclear Pt signal detected at dose of $1-100\mu$M. However, significant Pt signals were seen dose at $1\text{mM}$. The Pt image quality by micro-PIXE was considered adequate if both the nucleus and cytoplasm could be visualized well enough to allow performance of the procedure. The stronger signals were detected in the intracellular nucleus rather than in the cytoplasm. For time course study, longer exposure time of CDDP shows more uptake of the drug in the cells. The elemental analysis using air micro-PIXE can be applied for biomedical samples. Intracellular distribution of CDDP can be visualized with very fine resolution, and individual intracellular concentration of CDDP can be measured using our system.

シスプラチンは現在、頭頸部腫瘍から肺癌、消化器癌、婦人科癌にまで幅広い領域でkey drugとなっている。また、シスプラチンはこれらの腫瘍に対し、放射線治療と併用して、有効な増感効果をもたらすとされている。しかし、臨床ではシスプラチンによる治療効果には、患者ごとに明らかな感受性の差がある。もし、個々の癌細胞の取り込んだシスプラチンが画像化、定量化できれば、シスプラチン感受性やシスプラチン耐性機構の解析のために、重要な情報をもたらすと考えられる。一方、大気マイクロPIXE（Particle Induced X-Ray Emission）は数MeVに加速したプロトンを試料に照射し、放出される特性X線を検出することにより、試料中に含まれる多くの元素を$1\mu$mの分解能で解析できる装置である。個々の癌細胞の取り込んだシスプラチンを画像化、定量化できれば、シスプラチン感受性やシスプラチン耐性機構の解析において、重要な情報をもたらすと考えられる。

Fig.1. Three standard samples with different CDDP concentration was analyzed by in-air micro-PIXE

Fig.2. Intracellular and nuclear platinum count / nC according to CDDP exposure time

大気マイクロPIXEによるPtの定量性を確認するため、標準試料（Pt濃度を変化させた寒天を20μmに薄切し凍結乾燥したもの）を作成し、測定を行った。細胞はヒト肺腺癌細胞であるA549を用いて、$10\mu$MのBrdUでパルスラベルした対数増殖期の細胞を$5\mu$m厚のマイラーフィルム上で培養し、生着したことを確認した後、1mMのシスプラチンを接触させた。接触時間は30分から24時間とした。薬剤に接触後、THAMバッファーで6回洗浄した後、急速凍結乾燥した。解析は日本原子力研究開発機構高崎量子応用研究所の大気マイクロPIXE測定装置を用いた。

標準試料を用いた解析では、試料内のシスプラチン濃度と測定されたPtの間に直線性が明らかであった（Fig.1）。培養細胞試料では、P、K、Clなどの細胞の局在を示す元素、BrdUでラベルした核内Brおよび細胞に取り込まれたPtが検出可能であった。シスプラチン接触時間の延長とともに、細胞内Ptおよび核内Ptのカウントの増加が観察された（Fig2）が、生物実験に応用可能なPt濃度での検出は困難であった。

大気マイクロPIXEによるPtの細胞内可視化、定量化が可能であった。生物試料に応用可能なPt濃度と検出感度をあげる工夫が必要と考えられた。
3-67 Analysis of Trace Metal of HCV Replicon by In-air Micro-PIXE and ICP-MS

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We had reported that zinc was a negative regulator of HCV replication. The distribution of trace metal in HCV replicon (HCV-O) using in-air micro-PIXE and ICP-MS was analyzed. In-air micro-PIXE revealed more zinc in nucleus than in cytoplasm and no significant difference was observed in HCV-O and control (Huh7). On the contrary, ICP-MS proved that zinc bound-metallothionein was higher in HCV-O than in be needed to elucidate this discrepancy.

1. はじめに
我々は微量金属の亜鉛がC型肝炎ウイルス(HCV)に対して一定の抗ウイルス効果を有する事を報告してきた1,2)。しかし、亜鉛のHCVに対する作用は十分解明されていない。今回我々はin vitro HCV産生細胞であるHCV repliconを用い、micro PIXE及びその他の定量系によりHCVと微量金属、特に亜鉛の関係を調べた。

2. 実験方法
HCV replicon(HCV-O)は岡山大学加藤宣之教授より供与された。本細胞は人肝癌細胞株Huh7にfull lengthのHCVを感染させ樹立された。Huh7をコントロールとして、無刺激、Zn、interferon (IFN)、両者の併用による影響をみた。1) micro PIXEを用い細胞内微量金属を測定した。2) ICP-MSにより細胞内Zn、Cu、メタロチオネインの濃度を測定した。3) メタロチオネインは免疫染色、CdHem assayによっても測定した。

3. 結果
1) micro PIXEによって得たPとBrの分布から、細胞、核のcontourを同定した。さらにそれに基づき細胞のZnの分布をHCV-OとHuh7で比較したところ差は無く、Zn、IFN、両者の併用でも著明な差は見られなかった(Fig 1)。2) ICP-MSによる測定では、無刺激状態ではHCV-OはHuh7に比しZnは少ないが、Zn刺激によってHuh7より高まり、それはmetallothionein (MT)結合型であった(data not shown)。3) MTの染色では、いずれも亜鉛添加によりコントロールに比しMTは増加したが、HCV-Oでより強く増加した(Fig 2)。IFNと亜鉛併用でその傾向はより強くなった。

4. 考察
micro PIXEによる細胞個々の微量元素の分布をHCV-Oにおいて測定したのは今回が始めてであり、Znの核への局在は確認された。しかし、Huh7との違い、刺激による変化を観察することはできなかった。微量定量系であるICP-MSではZn刺激によりHCV-OはMT結合型のZnをより多く、細胞内に取り込むことが判明し、MT染色でも確認された。定量的にはHCVは明らかにZnの動態に影響を与えているが、micro PIXEではその違いを捉えられないとする結果であった。今後はmicro PIXEでの定量の可能性と、Zn変化のメカニズムについて検討したい。

References
3-68 Improved Radiosensitive Liquid-core Microcapsules for Targeting of Chemotherapeutic Agents

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We have reported that microcapsules consisting of alginate and hyaluronic acid can be decomposed by irradiation, and releases their core contents. These microcapsules have been generated by spraying a mixture of alginate and hyaluronic acid into CaCl₂ solution for more than 30 min. However, the use of Ca results in the hardening of the alginate, and the long polymerization time of more than 30 min results in the formation of microcapsules with thick outer shells. These factors prevent the efficient release of core contents from microcapsules.

During microcapsule development, we observed that yttrium (Y) prevents the regular polymerization of alginate by calcium (Ca), thereby allowing easy rupture of the microcapsules. Moreover, sonication of a mixture of alginate and hyaluronic acid results in shorter polymerization time that in turn results in the formation of microcapsules with thin outer shells. In this study, the effect of Ca-Y polymerization and sonication of materials were tested, using micro-PIXE camera.

A 0.2% (w/v) alginate solution and 0.1% (w/v) hyaluronic acid were mixed in an agar pestle; these percentages were determined in our previous experiment. To this mixture, 0.2 mmol carboplatin (a platinum (Pt)-containing anticancer drug) was added. A droplet of this mixture was sonicated using an ultrasound disintegrator and atomized to yield a 4.34% solution of CaCl₂ that was supplemented with Y at concentrations ranging from 0% to 0.01%. Resulting microcapsules was irradiated with single doses of 0.5, 1.0, 1.5, or 2 Gy ⁶⁰Co γ-radiation. Then a frequency of microcapsules’ rupturing were tested by micro PIXE camera.

The distribution of the microcapsular core contents (carboplatin) was imaged using a micro PIXE camera that detected platinum (Pt) signals (Fig. 1 a–h). The contours of the microcapsules were identified by detecting the distribution of Ca and ion beam-induced charge (IBIC) (Fig. 1(b), (d), (f), and (h)). Prior to irradiation, the microcapsule contours were clearly detected, and Pt was observed to be distributed within the capsules (Fig. 1(a), (b), and (d)). After irradiation, microcapsules with unclear Ca or Y distribution and with heterogeneous distribution of Pt (Fig. 1 (a)–(c) and (e)–(g)) were observed. We determined that these types of microcapsules had released their core contents upon irradiation. The frequency of microcapsules’ rupturing were shown Fig.-2. The increment in the frequencies of microcapsule decomposition was dependent on the final concentrations of the added Y (Fig. 2). When the concentration of the added Y exceeded 3.0 × 10⁻³%, the frequency of decomposition increased significantly (Fig. 2). Significant increases in these frequencies were also observed when the radiation dose exceeded 1.0 Gy (Fig. 2). The maximum decomposition frequency was observed when the final concentration of the added Y was 5.5 × 10⁻³% with 2.0-Gy irradiation. 2.0-Gy is the dose that is used in the conventional fractionated radiotherapy. Therefore, our microcapsules have a great possibility to be used in combination with clinical radiotherapy.

References
2) S. Harada IJ PIXE In press.
Fluorine Uptake into Enamel around Fluoride-containing Materials during pH-cycling

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Introduction
It is well established that the fluorine (F) is a possible agent for caries prevention. Also, the fluoride that contained some restorative material was demonstrated as a great benefit for preventing dental caries. The purpose of this study was to determine fluorine (F) uptake into enamel around fluoride-containing materials during pH-cycling using proton induced gamma-ray emission or PIGE technique.

Material and Methods
Class I cavities in the buccal surfaces of 6 extracted human teeth were drilled and filled with fluoride-containing materials; “Fuji IX (GC)” (FN) and “UniFil flow (GC)” with “MEGA bond (KURARAY)” (UF), and nonfluoride-containing material; “SOLARE (GC)” (SO). Four 300-µm sections through the material were obtained from each tooth. All specimens were polished to a thickness of 120 µm. All tooth surfaces, except outer surface of enamel, were coated with a wax. In order to simulate daily acid challenges occurring in the oral cavity, the pH-cycling (pH 7.0-pH 4.5) was carried out for 0, 1, 3 and 5 weeks, separately. After pH-cycling, F distribution of the outer lesion in each specimen was evaluated using PIGE technique. The amount of F in the outer 125 µm of the lesion was calculated. Two-way ANOVA and Bonferroni test were used for the analysis (p<.05).

Results
F distribution of the specimens clearly showed the F uptake from FN into only enamel adjacent to the material, while the F uptake from UF and SO were not detected. The enamel adjacent to only FN remained the area as caries inhibition zone in wall lesion. For FN, the amount of F uptake into the outer lesion increased during pH-cycling (Fig. 1). The amount of F uptake in 5-week pH-cycling had significantly higher value comparing to those in 1- and 3-week pH-cycling. For UF and SO, there were no significant differences among the duration of pH-cycling (Fig. 2 and 3).

Discussion
For fluoride-containing UF, the F uptake into cavity wall was not detected. This indicated the bonding layer between the tooth and fluoride-containing material interfered with the F absorption into the tooth. Therefore, there was not the area as caries inhibition zone in wall lesion. It was confirmed that the amount of F uptake from FN increased during pH cycling. In a pH of 5.5-4.5, hydroxyapatite dissolves and, by supplying fluorine, precipitation of fluorapatite (FA) occurs because of the continuing supersaturation of FA. This suggests that FN operated as a resource for supplying fluorine when the pH was in the low range.

Conclusion
Among fluoride-containing materials, there existed some differences in the F uptake with increasing pH cycling.

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H. Seito, T. Ichikawa, N. Haneda, H. Kaneko, Y. Sato,
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SiC-based Membrane for Gas Separation Synthesized by EB Curing of Precursor Polymers

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1. Introduction

SiC thin layer ceramics are expected to surpass the performance of polymeric, silica and carbon membranes. Beside superior mechanical behavior, they exhibit high oxidation resistance. Production of SiC coatings usually includes techniques where the substrate component is an inorganic SiC. However, employment of organometallic polymer precursors, such as polycarbosilane (PCS) deposited from solution followed by pyrolysis is a simple and convenient method for SiC production. Recently we applied PCS and/or polyvinylsilane (PVS) derived SiC-base coatings on γ-Al₂O₃ substrate, as membranes for gas separation. Nevertheless, thermal stability of the substrate limits pyrolysis temperature to relatively low, 850°C. Present report describes combination of two deposition methods, i.e. pulsed laser deposition (PLD) and polymer derived coating.

2. Experimental Procedure

α-Al₂O₃ was used as a substrate. Initially, PLD system was used with a SiC target at argon atmosphere and 500°C. Subsequently, a blend of PCS (Mn 2600) and PVS (Mw 2800, Mn 960) in cyclohexane solution was spin coated onto PLD surface. Samples were cured (or not) by EB, typically to dose of 1.2 MGy at a dose rate of 1.2 kGy/s, under dry air atmosphere. In order to transform the polymeric coating into an inorganic SiC-based one, samples were pyrolyzed in Ar atmosphere up to 850 °C using controlled heating ramp. Nitrogen and hydrogen gas permeation tests were conducted with samples of effective permeation area of 1.0 cm² by a vacuum method.

3. Results and Discussion

While considering fabrication methods of membranes for selective gas permeation, controlling of pore diameter is the crucial issue. Surface of PLD and PCS/PVS (80/20wt%) derived SiC deposited on alumina substrate is presented in Fig. 1. A rough surface resulted from PLD deposition (5h) does not provide satisfying membrane parameters: N₂ permeance was measured as 2.5x10⁻⁷ mol⋅sec⁻¹⋅m⁻²⋅pa⁻¹ and H₂/N₂ separation of 3.7 (Knudsen diffusion).

However, it provides proper support for polymer deposition. γ-alumina substrates with PLD originated SiC coated by preceramic polymer after pyrolysis shows relatively smooth surface. Thus we expect that such two-step technology of membrane preparation will be beneficial for its performance. Permeance of hydrogen and nitrogen was measured, and it was in the range 10⁻¹⁰ - 10⁻⁸ mol⋅sec⁻¹⋅m⁻²⋅pa⁻¹ depending on fabrication parameters. Accordingly, H₂/N₂ perm-selectivity was usually better than that characteristic for Knudsen diffusion. As an example, H₂/N₂ separation is presented in Fig.2. It was found nearly independent of PLD time, but strongly dependent on the polymer derived film. Here, one and two coating, irradiation, firing sequence is shown. Separation ratio is better, about 20, for twice coated substrate. So far, this separation ratio is smaller than that measured for membranes fabricated by four times polymer deposition. Therefore the combination technique presented here should be further developed.

References


Fig.1. Surface of (a) PLD and (b) Polymer derived SiC.

Fig.2. Perm-selectivity of N₂ and H₂. PLD followed by polymer deposition: PCS 80/20 PVS, 3% cyclohexane, spin coated at 1000rpm, irradiated at 1.2 MGy, fired at 850°C in Ar.
4-02 Effect of Ion Beam Irradiation on SiC Thin Film for Gas Separation

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1. Introduction

SiC thin film synthesized from Si-based polymer as a ceramic precursor by firing has an excellent thermal and corrosion resistance, and the structure is amorphous with nano-hole which has gas permeability. Therefore, the SiC thin film is expected as a hydrogen separation filter that can be used in the severe environment. However, the improvement of the amount of the hydrogen penetration and the separation ratio is required. In this research, the possibility of controlling the gas penetration of the SiC thin film was examined by the methods of ion implantation to form the hydrogen catalyst layer in the SiC film and of making nano-hole structure by passing high energy ions.

2. Experimental Procedure

The polycarbosilane (PCS) concentration of 2.5% in cyclohexane solution was spin coated at 1000 rpm onto porous alumina substrates, and the coated substrates were cured by thermal oxidation at 453K, and then fired in Ar at 1023K to transform the polymeric film into an inorganic SiC ceramic-based one. The SiC thin film with 0.5μm thickness was synthesized by repeating these process five times. Subsequently, the hydrogen and nitrogen permeation of the samples at 373 and 473K were measured by differential pressure method. The samples exhibiting the gas separation function by the molecular sieving mechanism were irradiated with several kinds of ions with energies enough to pass through the SiC thin film. After irradiation, nitrogen and hydrogen gas permeation tests were conducted with samples exhibiting effective permeation. Moreover, the SiC thin films of 0.2 and 0.4 μm in thickness were formed on the silicon substrate using similar process, and the change in the film thickness by the ion beam irradiation was examined.

3. Results and Discussion

Figure 1 shows the permeation ratios of hydrogen and nitrogen at 573K before and after irradiation of various ions at 1x10^{12}ion/cm^{2}. The permeation of nitrogen was not changed significantly by ion irradiation, but that of hydrogen decreased in 0.8 or less compared with that before irradiation, and the separation ratio of hydrogen/nitrogen became small. Moreover, when the ion fluence was increased up to 1x10^{14}ion/cm^{2}, the nitrogen permeation also decreased to 0.9 compared with un-irradiated samples.

Figure 2 shows the thickness change of the SiC thin film according to the ion irradiation of Ni 15 MeV. The thickness decreased over 20% at 1x10^{15} ion/cm^{2}. It is likely that the decrement of the film thickness was in proportion to the initial thickness. Therefore the surface of the film was not reduced by ion sputtering, but by the shrinking of the film in the thickness direction. The SiC thin film has amorphous ceramic structure fired at low temperatures compared with ceramic structural materials such as SiC fiber, and thus it has shrunk easily by ion beam irradiation as well as shrinkage of the amorphous silica film. The size of the nano-hole in which only hydrogen penetrates by the molecular sieve mechanism is about 0.3nm, and the number of the holes depends on the density of the SiC films. Therefore, the number of holes decreases by shrinkage, leading to a decrease in the amount of the gas permeation.

In the case of 0.5 MeV Pt irradiation, the projected range was estimated to be 0.14 μm below the SiC film thickness, and therefore the shrunk part was less than those of the other samples irradiated by high energy ions. However, the Pr irradiated film showed the smallest values of hydrogen and nitrogen permeation. According to the results of Rutherford Backscattering Spectrometry, the thin layer containing Pt was formed in the SiC film. It suggests that the formation of such thin layer reduces the number of nano holes, leading to the decrease of hydrogen and nitrogen permeation. Further investigations are necessary to clarify the mechanisms behind Pt irradiation.

References

![Fig. 1. Change in the hydrogen and nitrogen permeation ratios at 573K after irradiation of a variety of ions at 1x10^{12}ion/cm^{2}.](image1)

![Fig. 2. Thickness change of the SiC thin film according to irradiation of Ni 15MeV.](image2)
4-03 Formation of Si Nano-crystals and Control of the Spatial Distribution in Si-thermal Oxides


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Si nano-crystals attract much interest because of their unique electrical and optical properties. The formation of Si nano-crystals and control of spatial distribution are important in industrial devices. In this work, Si nano-crystals are fabricated in Si-thermal oxide film by ion implantation and thermal annealing. We have measured the dependence of the size and photoluminescence (PL) spectra of Si nano-crystals on the implantation doses. With increasing Si doses, the size of Si nano-crystals and the intensity of PL are increased, and the peak energy of PL is red shifted. This result indicates that size and PL of Si nano-crystals are controllable by Si implantation dose.

Si nano-ナノ結晶は量子サイズ効果による特異な電気的特性と光学的特性を持つため、多くの注目を集め、光・電子デバイスへの適用が検討されている。特に、その発光性や電荷蓄積機能を利用したエレクトロルミネッセンス(EL)素子およびメモリ機能への適用は多くある。半導体ナノ結晶は、自然界に大量に存在するため、環境負荷が小さい。半導体ナノ結晶は、産業用途においても重要である。本研究の目的は、シリコンナノ結晶の形成と、その空間的な配置制御である。そのため、シリコン熱酸化膜(膜厚600 nm)に対してシリコンイオン注入(Si+, 能量200 keV, 照射量:3-7×10^{16} ions/cm^2)を行い、シリコンナノ結晶形成のためのイオン注入条件を検討した。さらに空間的配置制御のため、電子あるいはイオンビーム照射によるシリコンナノ結晶析出が可能なシード基板の作成条件についても検討した。

実験では、窒素雰囲気中、1050℃において1時間の熱処理を行い、熱酸化膜中にシリコンナノ結晶を凝集させた1)。その後IFエッチングにより試料の表層300 nm程度のSiO_2を除去した後、原子間力顕微鏡(AFM)による表面観察を行った。また、顕微フォトルミネッセンス(PL)による光学特性評価(室温、488 nm励起)を行った。イオン注入後のシリコン熱酸化膜を熱処理後にエッチングし、AFMにて表面状態を観察した。イオン注入していない試料では、表面に析出物は観察されなかった。イオン注入した試料では、エッチング後に試料表面にシリコンナノ結晶の析出が観測された。イオン注入量に対するシリコンナノ結晶の高さ及び幅をFig.1に示す。注入量が増加するにつれて析出するシリコンナノ結晶の高さと幅が共に増大した。Fig.2にイオン注入及び熱処理後の試料のPLスペクトルを示す。イオン注入量の増加と共に、PL強度が増大し、ピーク波長は600 nmから690 nmにレッドシフトした。発光強度の増大はFig.1に示すシリコンナノ結晶の粒径の増大と共に、発光種が増加したためと考えられる。それと共に、量子サイズ効果あるいは界面の変化が生じ、発光種が変化することで、ピーク波長がシフトしたと考えられる。

電子ビームあるいはMeVイオンビーム照射効果を利用したシリコンナノ結晶析出のためのシード基板をシリコンイオン注入にて作製した。熱処理により凝集したシリコンナノ結晶をAFM及びPL測定により観察し、作製した試料がシード層として利用可能であることを確認した。シリコンナノ結晶の大きさとPLスペクトルの強度は注入量の増加と共に増大し、ピーク波長はレッドシフトした。この結果から、注入量を制御することでシリコンナノ結晶の大きさと発光を制御できることが確認できた。今後は、本シード基板に対して電子線や高エネルギープロトンビームの照射を行い、局所的にシリコンナノ結晶を析出させる予定である。

Reference
We have developed silicon carbide (SiC) coating for hydrogen gas separation membrane from a polymer blend of polycarbosilane (PCS) and polyvinylsilane (PVS). As one of the processes for synthesis of SiC coating, the PCS-PVS polymer blend is coated on the substrate, cured by irradiation in air, and pyrolyzed in an inert gas atmosphere. It has been found that the PCS-PVS polymer blend can be cured by the irradiation in air with dose as half as pure PCS. However, the mechanism of radiation oxidation curing of the PCS-PVS polymer blend has not been cleared. In order to clear the mechanism, it is useful to investigate the radical behavior of the PCS-PVS polymer blend because the radicals cause the oxidation and/or crosslinking of the molecules. In this work, the radical behavior of the PCS-PVS polymer blends was investigated.

Figure 1 shows the chemical structure of PCS and PVS. The PCS is a solid polymer at room temperature and its number average molecular weight is 2000. The PVS is a viscous liquid polymer at room temperature and its number average molecular weight is 960.

The PCS-PVS polymer blends were irradiated by gamma rays with dose of 0.2 MGy at liquid nitrogen temperature (77 K) in vacuum, and then the radicals in PCS-PVS polymer blends were measured by an electron spin resonance (ESR). The ESR measurements were also conducted for the PCS-PVS polymer blends irradiated by gamma rays with dose of 3 MGy at room temperature in air.

Figure 2 shows the relationship between the radical concentrations in PCS-PVS polymer blends and the PVS content after the irradiation of gamma rays at 77 K in vacuum. The radical concentration decreased linearly with increase in the PVS content. This suggests that the PVS does not affect on the production of the radicals in PCS-PVS polymer blends. Figure 3 shows the relationship between the radical concentration in PCS-PVS polymer blends and the PVS content after the irradiation of gamma rays at room temperature in air. Comparing with the result in Fig. 2, the radical concentration in the PCS-PVS polymer blends irradiated in air was low by a factor of $10^2$. This means that the produced radicals were decayed during the irradiation at room temperature in air. Furthermore, a tendency that the radical concentrations in PCS-PVS polymer blends were below the straight line that links the points of 0 % PVS and 100 % PVS was shown.

The decay of radicals in case of the irradiation in air is considered to be attributed by the recombination of the radicals (partly crosslinking) and production of peroxides (-OOH). So, the radicals in the PCS-PVS polymer blends may decay easily due to characteristics of the PVS such as liquid, low molecular weight, and rich Si-H bonds, that is, high mobility of molecules and high reactivity of Si-H bonds with oxygen. However, we can not distinguish clearly the recombination of radicals or the production of peroxides as a reason to cause the decay of radicals for the moment. Further investigation is necessary to know the radiation effects on the PCS-PVS polymer blends in detail.
Possibility of Silicon Carbide Micro-tube Synthesis from Polycarbosilane-silicone Oil Blend Precursors

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A blend polymer of 85 mass% polycarbosilane (PCS) and 15 mass% poly-hydro methylsiloxane (H-oil) was prepared as ceramic fiber precursor. Viscosity measurements on the blend polymer indicated that the blend polymer was sufficiently melt-spinnable at temperatures of 543 and 573 K. Optical microscope observation indicated that the fiber spun at 543 K was turbid, while the fiber spun at 573 K was transparent. IR spectra revealed that HS Low fiber (PCS – H-oil blend fiber melt spun at 543 K) maintained original H-oil content during melt spinning process, while the H-oil content in HS High fiber (PCS – H-oil blend fiber melt spun at 573 K) decreased. After thermal oxidation or radiation curing and pyrolysis at 1273 K, fibers with continuous pore structures were obtained. Continuous pore structure depends on spinning and curing conditions.

Fig. 1 on a sample, PCS was mixed with Si-H base containing type silicone oil and the blend polymer was heated to observe the viscosity change. The viscosity decreased by heating, but the temperature at which the viscosity started to decrease shifted to a lower temperature. This indicates that the silicone oil acts as a softening agent that partially dissolves in the PCS. At 543K, the fiber had a white opaque appearance, indicating that not all the oil was dissolved. The dissolution limit was estimated to be around 10 mass%.

At a lower spinning temperature, a small oil-rich domain was left, and then it was cured with an electron beam. The cross-sectional observation showed that there were submicroscopic pores in the form of a crescent shape. However, since radiation curing also affects the silicone oil, cases without pores were also observed, indicating the need for careful selection of radiation dose.

Low temperature spinning resulted in a decrease in the oil-rich domain, allowing the synthesis of silicon carbide fibers by electron beam curing without affecting the fiber. After curing, the fiber was observed to have a cross-sectional view showing a crescent-shaped pore. The oil content in the PCS polymer and the oil-rich part was measured to be around 10 mass%, indicating that the silicone oil had a partial dissolution in the PCS.

Fig. 1 Relative viscosity of PCS-H-oil polymer blend with temperature rising.

Fig. 2 Small crescent pores observed on the cross section of HS15 Low fiber cured with electron beam curing for 1.5h.
Effects of Ion Irradiation on Microstructural Change of SiC Nanotubes

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Since the discovery of carbon nanotubes (CNTs) in 1991, significant numbers of researches have prepared new one-dimensional nanostructured materials such as nanotubes, nanorods and nanowhiskers for potential applications. Some of them report that many nanomaterials such as TiC, NbC, BN, C/C/N, SiO\textsubscript{2} and GaN nanostructures are synthesized from CNTs as the template materials. We reported that the C-SiC coaxial nanotubes, which were CNTs sheathed with SiC, were formed. Furthermore, the single-phase SiC nanotubes were successfully synthesized by heating the C-SiC coaxial nanotubes in air. SiC is the most important semiconductor and has the excellent mechanical properties at high temperature. These SiC nanotubes are expected to be electrical devices and reinforcement materials used in radiation environments due to their excellent stability against radiations. Until now, there have been many researches about the effects of irradiation on the microstructural change of SiC bulk materials. However, the effects of irradiation on the microstructural change of the SiC nanostructure materials with peculiar shape such as SiC nanotubes in this study have not been investigated. The purpose of this study is, therefore, to investigate the effects of ion irradiation on the microstructural change of SiC nanotubes.

CNTs (GSI Creos Corporation, Tokyo, Japan) as the template materials and Si powder (The Nilaco Corporation, Tokyo, Japan) were used. The C-SiC coaxial nanotubes were synthesized by the reaction CNTs with Si powder at 1200 \degree C for 100h in a vacuum. Single-phase SiC nanotubes were formed by the heat treatment of C-SiC coaxial nanotubes at 600 \degree C for 2h in air. Thin films of single-phase SiC nanotubes were prepared by embrocating with the single-phase SiC nanotubes dispersed in ethanol on the alumina plates. These thin films of single-phase SiC nanotubes were irradiated by 3.0-MeV Si\textsuperscript{2+} ions at 300 \degree C and 900 \degree C. The irradiation dose was 6.4x10\textsuperscript{20} ions/m\textsuperscript{2}.

The results of X-ray diffraction measurements revealed that no change was observed by the ion irradiation at 300 \degree C and 900 \degree C.

The XPS spectra of the C 1s bands for the SiC nanotubes before and after ion irradiation at 300 and 900 \degree C are shown in Fig.1. There are two peaks representing Si-C and C-C in all specimens before and after ion irradiation. The energy difference between the peaks representing Si-C and C-C decreased with increasing the irradiation temperature. This result indicates that the binding state of SiC nanotubes changed by the ion irradiation at higher temperature. Similarly, the results of XPS measurements of the Si 2p bands for the SiC nanotubes before and after ion irradiation indicated that the peak representing Si-C existed in all specimens. The peak representing Si-O\textsubscript{2} also existed in the specimen before ion irradiation. However, the peaks representing Si-O and Si-O\textsubscript{2} disappeared by the ion irradiation.

The TEM observation revealed that the SiC nanotubes before ion irradiation were composed of the SiC grains with the size of a few nm to a few ten nm. Several single crystalline SiC nanotubes were formed by the ion irradiation at 900 \degree C. The result can be explained by the assertion that the grain growth of SiC nanotubes was enhanced by the ion irradiation at such high temperature.

References
4-07 Fabrication of Blue-light Emitting Fused-silica Substrates by Using Si-ion Implantation

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Introduction

Various works on silicon (Si) based luminescent materials utilizing the quantum confinement effect, such as Si nanocrystal (Si-nc), have been reported. Typical fabrication methods of Si-ncs are co-sputtering of Si and SiO$_2$1,2), laser ablation3), and Si-ion implantation into SiO$_2$ plates or thermal oxidized Si wafers4,5). Pavesi et al. reported that their samples including Si-ncs formed by using Si ion implantation exhibited photoluminescence (PL) and a large optical gain of 100cm$^{-1}$ in a wavelength range from red to infrared after annealing at 1100 °C. The optical gain is nearly equal to that of III-V semiconductors. Therefore, it has recently been anticipated that light emission devices composed of Si-nc-containing materials could be realized.

In this work, we fabricated Si-nc including fused-silica substrates by using the Si-ion implantation method and annealing, and evaluated their PL properties.

Fabrication and Evaluation

Si ions were implanted into a fused-silica substrate (10 mm x 10 mm x 1 mm$^3$) at room temperature in TIARA. The implantation energy was 80 keV, and the implantation amount was 1 x 10$^{17}$ ions/cm$^2$4). The Si implanted substrate was cut into four pieces (0.5 mm x 0.5 mm x 1 mm$^3$) using a diamond-wire saw and they were annealed in air for 25 min at 1100, 1150, 1200, and 1250 °C by using a siliconit furnace. PL spectra were measured at room temperature with excitation using a He-Cd laser ($\lambda$=325 nm). A monochromator, a photomultiplier, and a lock-in amplifier were used in our measurements. Figure 1 presents the PL spectra of the four samples generated at room temperature. Blue PL spectra having peaks around a wavelength of 400 nm were observed from all the samples. The peak wavelengths of the blue PL spectra of the samples are almost the same in spite of the varying annealing temperature. In our experiments, the blue PL peak had a maximum intensity after annealing at 1200 °C, and the intensity was 4.2 times higher than that of the peak of the longer wavelength band of the sample annealed at 1100 °C.

Discussion

Liao et al. reported that a blue PL having a peak wavelength of 470 nm was observed from Si-ion-implanted SiO$_2$ films and stated that the blue-light emission may have originated from an oxygen-defect level in SiO$_2$. However, the emission origin seems to differ from that of our samples owing to the difference of the PL peak wavelengths. In addition to a blue PL peak, a PL peak around a wavelength of 800 nm in a wavelength range from red to infrared was observed for the sample annealed at 1100 °C. In our experiments, the Si-ion-implant conditions were almost the same as those reported in ref. 4 and the annealing temperature of 1100 °C is common to ref. 4, therefore it seems that Si-ncs are also formed in this sample. There may be a common origin of blue-light emission in these samples and Si:SiO$_2$ co-sputtered films3). The SiO$_2$ layer located at the interfacial region of Si nanoclusters and surrounding SiO$_2$ medium may contribute to such emission. Blue-light-emitting materials are expected to be useful as light sources for optical pick-up systems. Therefore, we are trying to optimize the conditions of Si-ion implantation and annealing to improve the emission intensity and evaluate optical gains of the Si-implanted samples.

References


Fig.1 Measured PL spectra. PL peaks around a wavelength of 400 nm were observed from all the samples. The longer-wavelength peak was reduced after annealing above 1150 °C.
4-08 Effect of Impurity Co-implantation on Hydrogen Surface Exfoliation

H. Iwata, M. Takagi and Y. Tokuda

SOI (silicon on insulator) fabrication has become an attractive technology for low power, low voltage and high-speed electronics. Much attention has been given to the hydrogen exfoliation method introduced by Bruel 1 in comparison with other SOI fabrication techniques. This method involves the micro slicing process of silicon by high-dose hydrogen implantation and has advantages of greater uniformity of thickness and higher crystal quality of the silicon layer than other processes 1,2. Although this unique and useful process has been extensively developed in industrial applications during the past few years, the fundamental phenomenon and the underlying mechanism are still not completely understood 3. Behaviors of exfoliation on the surface were mainly examined by in-situ microscopic experience. From these results, we attempt to explain the relation between the defects distributions and the exfoliation phenomena, and report the effects of impurities (boron or fluorine) co-implantation.

Hydrogen ion implantation was performed in p-type Cz-grown (100) oriented silicon. The implantation energy was 80keV and hydrogen dose was 8×10ⁱ⁶ H/cm². After the hydrogen implantation, boron ion was implanted with 140keV. The projection range (Rp) of boron is almost coincident with the Rp of hydrogen. These specimens were annealed at in-situ heating optical microscopy (OM) experience. Annealing was carried out till 700°C. Defects distribution was estimated using cross sectional transmission electron microscopy (XTEM). Exfoliated area of surface were evaluated by scanning electron microscopy (SEM) and OM, and the depth of exfoliation was obtained with scanning prove microscopy (SPM). The damaged layer caused by hydrogen ion implantation is located in the region from 600nm to 800nm depth from the surface. Increasing the doses of boron below 3×10¹⁴ B/cm², the thickness of the damaged layer became narrower. Bellow 3×10¹⁴ B/cm² dosage, the number of exfoliated pieces was proportional to the boron dose, and average exfoliated diameter was proportional to the hydrogen dose. The total exfoliated area was maximized at 3×10¹⁴ B/cm² dosage. Over the 3×10¹⁴ B/cm² dosage, the number of exfoliated pieces was inverse proportional to the boron dose, and average exfoliated diameter was inverse proportional to the hydrogen dose. The total exfoliated area was maximized at 3×10¹⁵ B/cm² dosage. However, from XTEM observation, long crack was observed in the damaged layer of as implanted specimen. Using this condition, exfoliation with no annealing was observed.

Fluorine ion was implanted with 300keV. The projection range (Rp) of fluorine is almost coincident with the Rp of 80keV hydrogen. After the fluorine implantation, the defect distributions were observed by XTEM.

In a fluorine dose range from 1×10¹⁶ F/cm² to 5×10¹⁶ F/cm², a thick amorphous layer is formed in the surface vicinity. However, no differences were seen in the thickness of amorphous layer.

In the case of H and F co-implantation (as shown in Fig.1), crystal layer was found in the surface side. After the 700°C annealing, it was found that the thickness of crystal layer was proportional to the hydrogen dose. The result that hydrogen has an important role of impeding the amorphization

References
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Fig.1 Damage layers obtained by cross-sectional TEM. (a): (H 1×10¹⁶ + F 1×10¹⁶) / cm² as implanted. (b): (H 3×10¹⁶ + F 1×10¹⁶) / cm² as implanted. (c): (H 8×10¹⁶ + F 1×10¹⁶) / cm² as implanted. (d): (H 1×10¹⁶ + F 1×10¹⁶) / cm² as annealed. (e): (H 3×10¹⁶ + F 1×10¹⁶) / cm² as annealed. (f): (H 8×10¹⁶ + F 1×10¹⁶) / cm² as annealed.
4-09 Improvement of Gasochromic Properties in Tungsten Oxide by Ion Irradiation II.

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Tungsten trioxide, WO₃, has gasochromic coloration in that the color changes from yellow to dark blue in hydrogen atmosphere. Fiber-optic hydrogen gas sensors using a gasochromic WO₃ film have been studied to support the safe utilization of hydrogen as a clean energy source for the environment in recent years. One of the subjects of the investigation in the WO₃ film is large extent of the gasochromic coloration for satisfying the specifications of hydrogen sensors. The gasochromic properties are considered to relate to oxygen vacancies or defects in the tungsten oxide. In our study, ion beam irradiations have been tried to make oxygen vacancies in WO₃ thin films, and a possibility of the improvement in the gasochromic coloration has been investigated.

WO₃ thin films with the thickness of 0.4 μm were prepared on quartz glass substrates by reactive rf magnetron sputtering method with argon and oxygen mixture. After the deposition, the films were annealed at the temperature of 400 °C during 1 hour. The films were confirmed to have crystal peaks by the measurement of x-ray diffraction. The crystal structure was assigned to be monoclinic plane of WO₃. To investigate the gasochromic coloration of the films, a reduction of the intensity transmitted 630-nm-wavelength light was measured with an exposing of 1 % hydrogen diluted with argon gas. The typical reduction of the intensity at the exposure time of 600 seconds, that is, the extent of gasochromic coloration, R₀, was 15 %. He⁺ irradiations were performed in the fluence range from 5x10¹⁴ to 1x10¹⁷ ions/cm² at room temperature using the ion implanter at JAEA/Takasaki. The incident energy was selected 40 and 350 keV. After the irradiations, the extent of gasochromic coloration, R, was measured with the same procedure to the case of R₀.

In the previous report with x-ray photoelectron spectroscopic analysis, 20 % of the tungsten atoms in WO₃ film changes to oxygen-deficient tungsten caused by the 350 keV He⁺ irradiation at the fluence of 1x10¹⁷ ions/cm². Here, it is supposed that the oxygen vacancies of 1.1x10²² number/cm³ are created at the fluence of 1x10¹⁷ ions/cm², as the theoretical density of oxygen atom in WO₃ is 5.7 x10²² number/cm³. In the case of 40 keV irradiation, the amount of the oxygen vacancy is expected to be 8.2-fold of that in 350 keV from a calculation with theoretical density of monoclinic WO₃, 7.29 g/cm³. Assuming that the amount of oxygen vacancy is proportion to the fluence of 40 and 350 keV irradiation, the oxygen vacancy dependence of the gasochromic coloration is shown in Fig. 1. The value of R increases with increasing of the oxygen vacancy. At the amount of oxygen vacancy from 1x10²² to 5x10²² number/cm³, which is equivalent from 1 to 9 % of oxygen atoms in the WO₃ film, R has the maximum to be more than 3-fold of R₀. However, in the range above 5x10²² number/cm³, R returns to R₀. From this result, it is concluded that the optimum number of oxygen vacancy for high extent of the gasochromic coloration in WO₃ films exists.

Ion irradiation to WO₃ films was found to improve their gasochromic coloration by introducing oxygen vacancies, and expected to achieve high performance of gasochromic hydrogen sensor based on the WO₃ films.

References
Structural Change of Gasochromic \( \text{WO}_3 \) Films by Hydrogen Incorporation

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Structural change of the gasochromic \( \text{WO}_3 \) films by hydrogen incorporation was investigated. \((0 1 0)\)-oriented monoclinic \( \text{WO}_3 \) films were prepared by a sputtering method on \( \text{SiO}_2 \) substrates. When the light green color of Pd coated \( \text{WO}_3 \) films turned to dark blue by exposure of 1% \( \text{H}_2\text{-N}_2 \) gas, the monoclinic \( \text{WO}_3 \) structure changed to a tetragonal structure. The results of elastic recoil detection analysis (ERDA) and Rutherford backscattering spectroscopy (RBS) revealed that the hydrogen concentration increased 0.2 H/W in the samples during hydrogen exposing. These results indicate that the coloration of the \( \text{WO}_3 \) by hydrogen gas is caused by incorporation of hydrogen, with formation of hydrogen tungsten bronze (\( \text{H}_X\text{WO}_3 \)).

Fig.1. XRD patterns of the oriented \( \text{WO}_3 \) films (a) as-prepared and (b) colored states.

Fig. 2. Hydrogen depth profiling in the oriented \( \text{WO}_3 \) films (a) as-prepared and (b) colored states.
4-11 Effect of Annealing Temperature of Palladium Oxide Films on Hydrogen Detection

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The fast detection of hydrogen leaks below the lower explosive limit (LEL) of 4% by volume ratio of hydrogen in air is an important technology. Hydrogen sensors based on electrical properties of metal oxides have been developed. However, a hydrogen sensor, which can safely detect at room temperature, with a low cost and lightweight, is desirable. Recently, the gasochromic materials, coloration by gases, have considerable promise as the optical hydrogen sensing films. Palladium oxide films coated with noble metal (Pd, Pt) catalysts were expected to have gasochromic coloration, since it is known to exhibit optical absorbance changes due to the reduction with hydrogen.

Palladium oxide films are prepared by thermal oxidation of polycrystalline Pd films deposited on quartz substrates using an rf magnetron sputtering. The Pd films were annealed at temperatures range from 300 to 900 °C in air. For the gasochromic coloration measurements, the transmittance of palladium oxide films coated with Pd catalyst (15 nm thickness) was measured using a spectrometer with a fiber optics at a wavelength of 645 nm from a light-emitting diode. To confirm the detection of hydrogen less than LEL, the samples were exposed in 1 % H$_2$/Ar flow of 100 sccm. The transmittance T at a wavelength of 645 nm was measured as a function of exposure time of hydrogen. T$_0$ is the transmittance at the beginning of the measurements. The normalized transmittance T/T$_0$ of the film with Pd catalyst is rapidly decreased down to 0.1 within 2 min. The result shows that the Pd catalyst on palladium oxide film causes gasochromic coloration for hydrogen at room temperature.

The dependence of gasochromic performance on annealing temperature of polycrystalline Pd films is shown in Fig 2. The T/T$_0$ of the films on quartz substrates was taken after exposure 20 min in 1 % H$_2$/Ar. The T/T$_0$ of the films is decreased with increasing annealing temperatures and reaches to a minimum at 600°C. This result indicates that the palladium oxide film with superior gasochromic performance for hydrogen was obtained by annealing at 600°C in air. It is found that the gasochromic coloration of palladium oxide films for hydrogen is strongly influenced by annealing temperature. The composition of the palladium oxide films with different annealing temperatures was examined by the RBS. Polycrystalline Pd films (64 nm thickness) were annealed at temperatures range from 200 to 600°C in air for 1h. The results of RBS indicate the Pd phase remains up to 200 °C and begins to oxidize at around 250 °C and then becomes completely oxidized at 450 °C with formation of PdO phase. Palladium oxide film with PdO phase obtained by annealing at 600°C in air shows the appropriate gasochromic performance for hydrogen detection.

![Fig. 1 Optical response of the coloration for the palladium oxide films in 1 % H$_2$/Ar. (a) with Pd catalyst and (b) without Pd catalyst.](image)

![Fig. 2 The dependence of gasochromic performance on annealing temperature of polycrystalline Pd films.](image)
Improvement of Hydrogen Storage Characteristics of Palladium by Ion Irradiation

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In previous studies, the induction of vacancy in palladium (Pd) was found to be effective for an increase in the hydrogen absorption rate\textsuperscript{1, 2)}. Regarding the hydrogen storage in metals, it was reported that the absorption concentration of hydrogen atoms and the hydrogen absorption rate depend strongly on the surface state of metals\textsuperscript{1)}. For the surface modification of materials, ion implantation with low energy is known to be a quite useful method. These facts give the possibility that the hydrogen absorptivity in Pd is improved by surface modification using ion irradiation.

In this study, we perform chromium (Cr) ion irradiation into Pd, and the hydrogen absorption rate of ion irradiated Pd was evaluated. Based on the obtained results, we discuss the correlation among ion irradiation, work function and hydrogen absorption in Pd.

The samples used in this study were Pd sheets (99.99 \% purity, 7.5 x 7.5 x 0.1 mm\textsuperscript{3}). Prior to ion irradiation, all samples were annealed in nitrogen atmosphere for an hour at 1173 K. Cr ion (Cr\textsuperscript{+}) irradiation onto the Pd samples was made at room temperature using the ion implanter at TIARA. The acceleration energy of Cr\textsuperscript{+} and the maximum fluence were 350 keV, and 1 x 10\textsuperscript{17} /cm\textsuperscript{2} respectively. The hydrogen absorption rate of the irradiated Pd was investigated using an electrochemical method. Pd was used as a cathode, and an Hg/HgO was used as the reference electrode. Details of the electrochemical measurements have been reported elsewhere\textsuperscript{3)}.

Figure 1 shows the hydrogen absorption curves of Pd samples before and after 350keV-Cr\textsuperscript{+} irradiation. With increasing Cr\textsuperscript{+} fluence, the hydrogen absorption rate increased. The sample irradiated at a dose of 1 x 10\textsuperscript{17} /cm\textsuperscript{2} exhibited a much higher rate than the others, and the hydrogen absorption rate of the Pd became five times higher than that of un-irradiated Pd. Since vacancy type defects are introduced by irradiation and such vacancies might act as hydrogen trapping sites, the enhancement of the hydrogen absorption rate obtained in this study can be interpreted in terms of an increase of hydrogen trapping sites near the surface region due to irradiation of Cr\textsuperscript{+} into Pd.

Figure 2 shows the relationship between the work function of Pd (\(\Delta\phi\)) and the hydrogen absorption rate before and after Cr ion irradiation. The value of \(\Delta\phi\) is depicted as a shift from un-irradiated sample. The shift of the \(\Delta\phi\) is estimated from \(\Delta\phi = \frac{V_E}{\Delta V - 1}\), where \(V_E\) is the peak-to-peak output voltage and \(\Delta V\) represents the voltage difference between the probe tip and Pd sample. By Cr\textsuperscript{+} irradiation, \(\Delta\phi\) shifts to the negative side. As for the hydrogen absorption rate, the value increased with increasing fluence of Cr ions. These results imply that the value of \(\Delta\phi\) decreases and the hydrogen absorption rate increases with increasing vacancy concentration.

In summary, Cr ion irradiation with energies of hundred keV is useful for the improvement of the hydrogen absorption rate of Pd.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Hydrogen absorption, 5 cycles, curves of Pd irradiated with 350keV Cr\textsuperscript{+} at doses from 1x10\textsuperscript{14} /cm\textsuperscript{2} to 1x10\textsuperscript{17} /cm\textsuperscript{2}.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{Relationship between the work function of Pd (\(\Delta\phi\)) and the hydrogen absorption rate before and after Cr ion irradiation.}
\end{figure}

References
4-13 Gamma-ray Irradiation Effect on Lattice Defect Centers Detected from the Tsurukawa Fault Gouge, Japan

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A well-known method for the assessment of fault activity (i.e., seismicity) is the ESR (electron spin resonance) dating method using lattice defect centers detected from intrafault material (fault gouge)\(^1\). Clay minerals such as smectites in fault gouge are considered to be produced at almost the same geological time as the fractures of host rocks during fault movements, so that ESR ages obtained from the clay minerals may directly show the ages of fault movements. In this report, we investigate \(\gamma\)-rays irradiation effect on ESR centers detected from the fault gouge along the Tsurukawa fault, which is distributed from Yamanashi to Kanagawa Prefecture, and consider whether or not the ESR centers can be used for ESR dating of fault movements. The experimental fault gouge samples were collected from an outcrop along the Kaisawa River, Tabayama Village, Yamanashi Prefecture. At this outcrop, a black fault gouge layer (T-3) has been formed along the fault plane that may have been most active at the latest fault movement, and dark gray gouge layers (T-2, T-4) are positioned on both sides of the black fault gouge. Furthermore, gray gouge layers (T-1, T-5) are distributed outside the dark gray gouge layers. The source rock of these fault gouge samples is pelite.

The \(\gamma\)-irradiation was carried out using a \(^{60}\)Co source in the 6 cell irradiation facility, Takasaki Advanced Radiation Research Institute, JAEA. The dose rate and time of irradiation are 23.7 C/kg/h (0.97 kGy/h) and 0~10 hours. ESR measurements were carried out using an X-band ESR spectrometer with a 100 kHz modulation field (JEOL RE-3X) in Yamaguchi University. As a result of ESR measurements, paramagnetic Fe\(^{3+}\) and Mn\(^{2+}\) ions signals are detected from all the fault gouge samples. In addition, ESR signals (g=2.0123, 1.9973) intrinsic to smectites, an E’ center (g=2.0015) derived from oxygen vacancies, an organic radical center (g=2.0039), and an unknown ESR signal (g=2.0041) are detected especially from T-3 or T-4 sample. Figure 1 shows variations of ESR spectra obtained from T-3 and T-4 samples before and after \(\gamma\)-irradiation. As shown in the figure, the ESR signals intrinsic to smectites regularly increase with \(\gamma\)-irradiation. This result means that the ESR signals of smectites are applicable to ESR dating of fault movements. On the other hand, the E’ and organic radical centers irregularly change by \(\gamma\)-irradiation. There have been hardly experimental data on the \(\gamma\)-irradiation effect on organic radical centers. Our result indicates that the organic radical centers are not available for ESR dating.

Furthermore, the unknown ESR center at g=2.0041 clearly increases with \(\gamma\)-irradiation, so that it may be applicable to ESR dating of fault movements. According to X-ray diffraction analysis, this unknown center may be derived from another clay mineral such as illite. To identify this unknown center, we need to carry out more detailed experiments such as comparison with ESR signals detected from standard clay minerals.

Reference

Provenance Study of Eolian Deposits Using Electron Spin Resonance Signal Intensity and Crystallinity of Quartz

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Inland desert areas of western China such as Taklimakan and Gobi are special for their distribution in higher latitudes, and it has been suggested that uplift of Himalaya and Tibet is a cause of their formation based on paleoclimatic model simulations. Thus, specification of the timing and mode of formation of these deserts is critical to test the potential linkage among the uplift of Himalaya-Tibet, the evolution of Asian monsoon and the formation of deserts in inland Asia. Then, we aim to reconstruct past provenance changes of eolian deposits in East Asia in order to examine the timing of formation of these deserts and the evolution of Asian monsoon. In order to estimate the provenance of eolian deposits, we focused on Electron Spin Resonance (ESR) signal intensity and crystallinity index of quartz, which are powerful methods to constrain on the provenance of dust deposits. Here we first determine the procedure of gamma-ray irradiation for ESR analysis.

References
4-15 Preparation of Anisotropic Conductive Membranes Using Ion Beam Irradiation

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The damaged region created along the path of swift heavy ions in polymers is susceptible to a proper etchant, resulting in the formation of through-holes with nanometer-to-micrometer diameters. This so-called ion track membranes of thermally and mechanically stable polymers have been developed for applications to electronic devices through hybridization with metals and semiconductor alloys by electroplating\(^2\). In this study, anisotropically conducting membranes were prepared by electrochemical deposition of copper (Cu) wires into the cylindrical pores in the polyimide (PI)-based ion track membrane. We report here the conductivity of PI/Cu composite films with low fluence Cu wires.

Commercially-available PI films (Kapton, thickness: 12 µm) were irradiated by 450 MeV \(^{129}\)Xe\(^{2+}\) with fluences of \(3 \times 10^3\) and \(3 \times 10^4\) ions/cm\(^2\) from the AVF cyclotron at the TIARA facility. According to the established procedure\(^3\), the PI ion track membranes with 0.5 – 2.0 µm diameter cylindrical pores were then prepared by the etching in an aqueous sodium hypochlorite (NaClO) solution at 60°C. Successive layers of gold (25 nm) and Cu (20 µm) were coated on one side of the ion track membranes as a cathode\(^4\). The Cu wires were deposited within the pores of the ion track membranes onto the Cu cathode by electrochemical plating in a pH 11 aqueous solution of 1.3 M copper sulfate (CuSO\(_4\)). The dot of Au electrodes with 0.3 - 0.5 mm in diameter was deposited on the other side of the Cu cathode to connect with copper wires of the PI/Cu composite films. The Au dot electrodes were connected to two terminals of milliohmeter via Au wires, then electrical conductivity of the PI/Cu composite films was measured.

The electric resistances of the ion track membranes with pores of 0.5 µm in diameter and a fluence of \(3.0 \times 10^5\) ions/cm\(^2\) were measured using the dot electrodes in 8 places on the PI/Cu composite films. The measured (B) and theoretical (A) resistances and these ratios (B/A) were shown in Table 1. Theoretical resistances were given by the next equation,

\[
R = \frac{1}{S} = \frac{\rho L}{\pi (d/2)^2 A F}
\]

where \(\rho\) is a resistivity of copper (1.67\(\times\)10\(^{-6}\) Ωcm), \(L\) is length of copper wires, \(d\) is a diameter of the copper wires, \(A\) is an area of the electrode, and \(F\) is a particle fluence.

Somewhat lower conductivity of the PI/Cu composite films compared with the theoretical value is likely to result from the defects in copper wires during the electroplating preparations, influence of the interfaces between copper wires and the pore surfaces of PI, and/or the contact resistances between the Au dots and the top of copper wires.

### Table 1 Resistance that calculated the area of each electrode and the magnification to the theoretical resistances of measurements

<table>
<thead>
<tr>
<th>Area of the electrode cm(^2)</th>
<th>Theoretical resistance, A m Ω</th>
<th>Measured resistance, B m Ω</th>
<th>Ratio (B/A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 2 x10(^3)</td>
<td>2.8</td>
<td>23</td>
<td>8.3</td>
</tr>
<tr>
<td>2 2.7 x10(^3)</td>
<td>1.3</td>
<td>7.2</td>
<td>5.7</td>
</tr>
<tr>
<td>3 2.3 x10(^3)</td>
<td>1.5</td>
<td>8.3</td>
<td>5.6</td>
</tr>
<tr>
<td>4 2.8 x10(^3)</td>
<td>1.2</td>
<td>11.8</td>
<td>9.8</td>
</tr>
<tr>
<td>5 3.1 x10(^3)</td>
<td>1.1</td>
<td>8.4</td>
<td>7.7</td>
</tr>
<tr>
<td>6 4.2 x10(^3)</td>
<td>0.8</td>
<td>6.9</td>
<td>8.5</td>
</tr>
<tr>
<td>7 3.5 x10(^3)</td>
<td>1.0</td>
<td>7.2</td>
<td>7.5</td>
</tr>
<tr>
<td>8 3.3 x10(^3)</td>
<td>1.0</td>
<td>7.2</td>
<td>7.0</td>
</tr>
</tbody>
</table>

Figure 1 The histogram of the distribution of 21 points of the measured conductivity in the composite films with the copper wires; fluence: \(3 \times 10^4\) ion/cm\(^2\), diameter: 2 µm.

Thus, we used the samples with low density of Cu wires (\(3 \times 10^3\) ion/cm\(^2\)) with low resistance of each wire (2 µm in diameter) in order to obtain the resistivity information of single wire.

Figure 1 shows the histogram of the distribution of the measured conductivity of 21 points of the PI/Cu composite films with pores of 2 µm and a fluence of \(3 \times 10^3\) ion/cm\(^2\). There was three distributions with peaks at conductivities of 15, 35 and 55 S with the equal intervals. These peak conductivities are in good agreement of the integral multiple of the theoretical conductivity of one copper wire of about 17 S. It was considered that the copper nanowires in the composite membranes formed by using the ion track films had the conductivity equal with the bulk. However, lower conductivity of the PI/Cu composite films with pores of 0.5 µm in diameter should results from some of the wires are not connected to the Au dot electrodes.

### References
Development of Optical System for Direct Observation of Behavior of Transient Species under Pulsed-heavy Ion Irradiation

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1. Introduction

High energy heavy ion irradiation induces characteristic chemical reactions in water, which are considered to be caused by high concentrations and heterogeneous reactions of radicals in track. Hydroxyl(OH) radical is the most important radical for oxidations in water. We reported the dependence of the differential $G$-values of OH radicals on the energy and mass of the incident ions, and on the elapsed time by the product-analysis method. In order to understand precisely the reactions in track, direct observation method is more useful and informative. We construct the optical system under the pulsed heavy ion irradiation.

2. Experimental

Carbon ion with 220-MeV energy was selected to evaluate and improve the specification of the constructed optical system. As shown in Figure 1, C ion beam from an ion source was bunched by a chopper to obtain the short pulse with a width of several $\mu$s to 1000 $\mu$s, which was controlled by a pulse generator. Time profile and charge of the pulse were directly measured by Faraday cup set at the upper stream of the beam in vacuum. Sample cell was horizontally set on the optical stage under the window for extracting the ion from vacuum. The sample cell has two glass windows with 50-$\mu$m thickness for the ion irradiation and optical measurement on the top and bottom. Probe light was introduced into the sample cell with an angle of 30 degree against the ion beam axis, and reflected by mirror under the cell. Light intensities observed by a photodiode with and without irradiation are defined as $I$ and $I_0$, respectively. Absorbance due to produced radicals is estimated by Lambert-Beer equation ($OD=\log(I/I_0)$). Improving optical system and reducing electric noise make possible to measure a small absorbance less than $10^{-4}$.

KSCN dissolved in water was used as sample solution, because its reaction mechanism is well known and primary radical has a large extinction coefficient.

3. Results and discussion

Time dependent absorbance was recorded at 447 nm for aqueous KSCN solution irradiated with C ion as shown in Figure 2. OH radicals produced in the track react with solute to produce (SCN)$^2-$, and absorbance shown in Figure 2 is assigned to (SCN)$^2-$. Rise time of absorbance from time 0 is completely the same as the irradiation pulse width. Increase in absorbance with pulse width implies that (SCN)$^2-$ is relatively stable and its concentration increases. Saturation observed for the longer pulse irradiation demonstrates the competition between the formation and decomposition of (SCN)$^2-$.

Reference


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Fig. 1 On-line optical measurement system for pulsed heavy ion irradiation.

Fig. 2 Time dependent absorbance for 10-mM aqueous KSCN solution irradiated with C ions having different pulse widths.
4-17 LET Effect on Irradiation of Hydroxymaleimide in Alcohol Solution

S. Nakagawa \textsuperscript{a)}, N. Ohta \textsuperscript{b)}, M. Taguchi \textsuperscript{c) and K. Hirota \textsuperscript{c)}

\textsuperscript{a)TMITRI, \textsuperscript{b)Hiroshima Univ., \textsuperscript{c)Environment and Industrial Materials Research Division, QuBS, JAEA}

Hydroxymaleimide was irradiated in 2-propanol and methanol solutions with 350 MeV Ne-ion, 220 MeV C-ion, 100 and 50 MeV He-ion beams from the AVF cyclotron in the TIARA Facility. The differential G-values of the reduction of hydroxymaleimide decreased with increasing LET. The LET dependence by He-ion shifted from heavier ions. This effect clearly appeared in methanol solution.

Let's analyze the text in detail:

**Title:** LET Effect on Irradiation of Hydroxymaleimide in Alcohol Solution

**Authors:** S. Nakagawa \textsuperscript{a)}, N. Ohta \textsuperscript{b)}, M. Taguchi \textsuperscript{c) and K. Hirota \textsuperscript{c)}

**Institutions:** \textsuperscript{a)TMITRI, \textsuperscript{b)Hiroshima Univ., \textsuperscript{c)Environment and Industrial Materials Research Division, QuBS, JAEA}

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**Discussion:**

- **Fig. 1.** Apparent G-value depending on the incident energy of ion in 2-propanol.
- **Fig. 2.** LET dependence of differential G-value obtained by ion irradiation in 2-propanol.
- **Fig. 3.** LET dependence of differential G-value obtained by ion irradiation in methanol.

Let's break down the main points:

1. **Hydroxymaleimide irradiation:**
   - Irradiated in 2-propanol and methanol solutions with various ion beams (Ne, C, He).
   - The differential G-values decreased with increasing LET.

2. **LET effect:**
   - The LET dependence by He-ion shifted from heavier ions, which was more evident in methanol.

3. **Experimental methods:**
   - Prepared 0.1 M solutions of hydroxymaleimide in 2-propanol and methanol.
   - Irradiated with Ne (350 MeV), C (220 MeV), and He (100 MeV, 50 MeV) ions.
   - Used Al foil to control LET increase.

4. **Analysis:**
   - Analyzed by liquid chromatography-mass spectrometry (ODS column, 50:50 acetonitrile-water).
   - Analyzed using ES-NCI ionization.

5. **Results and Discussion:**
   - Observed the decrease in hydroxymaleimide concentration with increasing LET.
   - The apparent G-value increased with the incident energy of ions.
   - The differential G-values for He ions showed a clear shift from heavier ions.

6. **Conclusion:**
   - Further studies are planned to investigate the relationship between LET and the formation of active species in the presence of additives.

This study highlights the importance of LET in the irradiation of hydroxymaleimide, demonstrating that heavier ions have a stronger effect compared to He ions, particularly in methanol. The findings contribute to the understanding of radiation chemistry and could have implications for the fields of radiation biology and materials science.
4-18 Transient Absorption of a Cation Radical of Pyrene Induced by Heavy Ion Beam Pulses

T. Kondoh a), J. Yang a), K. Kan a), Y. Yoshida a), H. Shibata b), M. Taguchi c) and T. Kojima c)

a) ISIR, Osaka University, b) Faculty of Engineering, Kyoto University, c) Environment and Industrial Materials Research Division, QuBS, JAEA

The heavy ion beam is a high LET radiation. Ion beam can excite materials high densely. The characteristic of this ion beam was used to develop the new technology such as the material and the medical application. Ion beam pulse radiolysis is developing for elucidation of the initial process of ion beam induced chemical reaction. Our unique measurement method is developing using a scintillator and the photon counting method. Transient optical absorption of the Pyrene’s cation radical which was induced by irradiation of 220 MeV C⁵⁺ ion was measured. A decay time of cation radical of pyrene in ion beam case was faster than that in electron beam case.

Fig. 2 Transient Optical absorption of 100mM Pyrene-CH₂Cl₂ solution at 440nm induced by Ion beam (Gray dots) and electron beam (Black triangle dots)

Fig. 1 Measurement system of ion beam induced optical absorption
4-19 Basic and Application Studies on Chemical Responses to Quantum Beams in Heterogeneous Systems

R. Nagaishi a), R. Yamada a), N. Aoyagi a) and Y. Sugo b)

a)Division of Environment and Radiation Sciences, NSED, JAEA, b)Division of Fuels and Materials Engineering, NSED, JAEA

From the standpoints of utilization of radioactive wastes, and of sophistication of separation process of spent fuels, we have been investigating promotion or inhibition of radiation-induced reactions in immiscible heterogeneous systems: solutions coexisting/contacting with solid oxides, solvent system with aqueous and organic phases, etc.. We have recently report that the reactions of reduction of metal ions and of hydrogen production in aqueous solution were promoted by adding oxide particles to the solution 1, and that the radiolysis of amides in n-dodecane was dependent on aqueous solution contacting with the n-dodecane 2). In this report, we illustrate recover of platinum-group elements from aqueous solution, and non-toxic treatment of chrysotile asbestos using ionizing radiations as the experimental results found in fiscal 2006.

The samples of Pt-group element ions and asbestos were irradiated at ambient temperature and pressure using the electron beams (initial energies: 0.5-0.8 MeV) and γ-ray (avg. one: 1.25 MeV) at JAEA-Takasaki: the dosimetry was made by using the CTA-film and/or solution dosimeters. After the irradiation, ions/molecules formed in the solution part of sample were measured by using spectrophotometry, liquid chromatography and ICP-AES; the gas part such as H₂ gas was produced from the solution (b): the yield of H₂ gas was produced from the solution (b); the yield in the presence of asbestos was much larger than those in the absence of asbestos or in the presence of oxide particles 3).

A large amounts of Pt-group elements, especially Pd, Re and Ru, were contained in high level wastes discharged from power reactors, and expected to be recovered from there. A large amounts of Pt-group elements, especially Pd, Re and Ru, were contained in high level wastes discharged from power reactors, and expected to be recovered from there. Figure 1 shows the radiation-induced reductions involving Pt-group elements. When considering the recover of elements by using radiations, Pd(II), Rh(III) and Pt(IV) ions were effectively reduced and solidified in the presence of a small amount of oxide particles without using reducing agents such as alcohol radicals, which can be formed by adding alcohols to the solution, although not in the absent (a); the solidified ones were dispersed on the particles and recovered. When using the recovered particles, Ce(IV) ion was reduced more effectively than the original ones.

Asbestos is discharged from general/industrial wastes in large amounts. Since it is so toxic due to its needle-shape and to its resistance to heat and chemicals, a novel method for the non-toxic treatment without using high-temperature heat and harmful chemicals is expected. Figure 2 shows results for asbestos irradiated in aqueous solutions. While the dried asbestos did not change before and after irradiated, its change from needle- to particle-shape (a) and dissolution into the solution rapidly proceeded with the increasing the absorbed dose and acidity in the solution when it was immersed into the solution. During the morphological, H₂ gas was produced from the solution (b); the yield in the presence of asbestos was much larger than those in the absence of asbestos or in the presence of oxide particles 3).

References


Fig. 1 The radiation-induced reductions involving Pt-group elements in aqueous solutions. (a) The recovering of Pd: when adding oxide particles adding to the solution, Pd(II) ion was effectively dispersed on the particles. (b) When using the dispersed particles, Ce(IV) was reduced more effectively than the original ones.

Fig. 2 Morphological change of needle-shaped asbestos and H₂ production in aqueous solutions. (a) The XRD peak at 24° indicating the crystalline asbestos decreased with increasing the dose and acidity. (b) The yield of H₂ produced in the presence of asbestos was much larger than those in the absence of asbestos or in the presence of oxide particles.
Effect of Ion-irradiation and Annealing on Pinning Property of PLD Prepared YBCO Tapes

K. Nakashima a, b, N. Chikumoto b, A. Ibi b, S. Miyata b, Y. Yamada b, T. Kubo c, A. Suzuki c and T. Terai c

a) Department of Quantum Engineering and Systems Science, The University of Tokyo,
b) Superconductivity Research Laboratory, ISTEC, c) Department of Nuclear Engineering and Management, The University of Tokyo

The change in pinning properties of YBCO tapes prepared by pulsed laser deposition (PLD) method was investigated after ion-irradiation and thermal annealing treatment in order to confirm that ion-irradiation is effective for the enhancement of pinning properties. In this study, YBCO tapes (2 mm×2 mm×0.5 mm) were irradiated with 400 MeV Kr$^{18+}$, 450 MeV Xe$^{23+}$ or 500 MeV Au$^{31+}$ from $1.0\times10^{10}$ to $1.5\times10^{12}$ ions/cm$^2$. As a result of ion-irradiation, the enhancement of $J_c$ was observed especially at high applied magnetic field in 77 K. After the ion-irradiation, the samples were annealed at the temperature of 473 K or 673 K in the oxygen atmosphere for 2 h. From the post-annealing experiment, it was clarified that oxygen vacancies and interstitial atoms in the irradiated YBCO tapes can be recovered at the temperature of 473 K and 673 K, respectively.

1986年に銅酸化物における高温超伝導が発見されて以来、液体窒素温度である77 Kにおける超伝導応用へ向けた種々的研究開発が行われている。HTSCは第2種超伝導体であるため、磁場中では量子化された磁束（Vortex）がHTSC内へ部分的に進入する。Vortexが超伝導電流により駆動されると電気抵抗が発生してしまうことから、Vortexをピン止めする必要が生じる。ピンニングセンターとなるものは超伝導体中の欠陥や不純物などであり、高い臨界電流密度($J_c$)を実現するための導入方法の開発が進められている。量子線照射は、製作過程によらず、照射条件を変化させることで超伝導体内のピンニングセンターの形状・密度を制御できる利点がある。そこで本研究においては、重イオンビーム照射を用い、現在開発が進められている超伝導線材であるPulsed laser deposition法（PLD法）により作成されたYBCO線材（Y系PLD線材）へ照射欠陥を導入し、$J_c$などの超伝導特性の変化を調べた。

照射用試料は(財)国際超電導産業技術研究センター超電導工学研究所で作製されたY系超伝導線材を2 mm×2 mm×0.5 mm程度に切り出して作製した。高エネルギー重イオン照射をAVFサイクロトロンを使用した。照射は保護層であるAg層（5 μm）側からYBCO層（0.5 μm）に垂直に照射した。照射イオン種はYBCO層を貫通する条件として、400 MeVのKr$^{18+}$、450 MeVのXe$^{23+}$、500 MeVのAu$^{31+}$を選択し、照射フルエンスは$1\times10^{10}$～$1.5\times10^{12}$ [ions/cm$^2$]の範囲とした。PPMS（Quantum Design社製）を用いた磁化測定により、転移温度($T_c$)、臨界電流密度($J_c$)等の評価を照射前後で行った。照射後アニールは、照射試料を酸素雰囲気中で目的の温度、時間で保持した後に銅板上で急冷し、その影響を調べた。

温度77K、印加磁場2Tにおける照射フルエンスと$J_c$の関係をFig.1に示す。どのイオン種でも$5\times10^{11}$ [ions/cm$^2$]程度のフルエンスで最も$J_c$が向上した。また、$J_c$が最も向上するイオン種はXeイオンであるが、これは他の理由は結晶性を大きく損なわずに有効な円柱状欠陥を多数導入できるためであると考えられる。

照射後アニール実験は473Kおよび673Kについて2時間行った。473 Kでは酸素欠陥が、673 Kでは酸素欠陥が、それぞれ過剰フルエンス照射の場合に特に大き回復することが分かった。

Damage Evaluation by Means of Electrical Resistivity Measurement in Oxide Irradiated with MeV Electrons

N. Ishikawa a), S. Yamamoto b), Y. Chimi c), H. Sugai a), T. Kato a) and A. Iwase d)

a) Division of Fuels and Materials Engineering, NSED, JAEA b) Environment and Industrial Materials Research Division, QuBS, JAEA, c) LWR Long-term Reliability Research Unit, NSRC, JAEA d) Department of Materials Science, Osaka Prefecture University

One of the well-known standards for describing radiation damage density in solids irradiated with high energy (keV-MeV) particles is called dpa (displacements per atom). Radiation damage created by different radiation conditions can be compared respectively with the common standard, if dpa is adopted. Especially when the target material is a simple metal, this evaluation method is widely used. This is because relatively simple defects are created in an irradiated simple metal which consists of one atom species. Moreover, elastic displacement is the only origin of the defect formation for simple metal, and this facilitates the estimation of the dpa value.

However, it is not established for oxide ceramics materials that this method can be simply applied for the estimation of damage density, and there is a significant possibility that complicated defect formation or annihilation can take place. Moreover, for oxide ceramics, the method for experimentally evaluating radiation damage has to be re-examined in the first place.

In this study electrical resistivity measurement is adopted for evaluating radiation damage in oxides as it is widely used for the evaluation of radiation damage in metals. Electron irradiation of TiO$_2$ and in-situ electrical resistivity measurement were performed. The energy range is restricted in the range of 1.0-2.5 MeV, since in this energy range the irradiation with electrons is known to cause elastic displacements of target atoms, and relatively simple defects are created.

Thin films of TiO$_2$ were deposited on SrTiO$_3$ (100) single crystal substrates, and were irradiated with 1.0 MeV and 2.5 MeV electron beam. The thickness of the films was about 0.3 μm which is thin enough for the electrons to pass through. The irradiations were performed at room temperature (280-283K), and the fluence dependence of electrical resistivity was measured in-situ at a fixed temperature of 280K by means of the conventional four probe method.

In Fig.1 the electrical resistivity as a function of fluence is shown for TiO$_2$ irradiated with 1.0 MeV and 2.5 MeV electrons. The monotonic decrease of the electrical resistivity is found for both irradiations in contrast to the monotonic increase found for high-energy ion irradiations. The decrease in electrical resistivity probably originates from increase in carrier concentration which may be related to displacements of oxygen atoms. For high fluence region (>1x10$^{15}$ cm$^{-2}$) the electrical resistivity can be described by the linear function of the logarithm of fluence. In this fluence range, the calculated atomic displacements are in the order of ppm, and the mutual interaction between created defects is not intense. Moreover, the change in mobility due to increase in scattering center for conduction electrons is probably very small.

The slopes of the curve shown in the figure are almost the same, and the defect creation cross section for 2.5 MeV electron irradiation is higher than for 1.0 MeV electron irradiation, indicating that the energy increase contributes to enhancement of defect creation cross section. The investigation over wider energy range is necessary to describe more precise energy dependence of the resistivity behavior.
4-22 Lattice Defects Induced by Electron Irradiation in FeRh Alloys

A. Iwase a), A. Ishii a), F. Hori a), Y. Zushi a) and N. Ishikawa b)

a)Department of Materials Science, Osaka Prefecture University,
b)Division of Fuels and Materials Engineering, NSED, JAEA

FeRh alloys were irradiated with 0.80-MeV and 2.0-MeV electrons, and the defect structure was studied by means of a positron annihilation technique. The experimental result shows that the electronic structure around irradiation-induced vacancy-type defects depends on the energy of electrons.

Fe-50%Rh is a B2-type compound, and the phase transition to the ferromagnetic phase occurs near room temperature. Recently, we have shown that FeRh becomes ferromagnetic even at low temperatures when irradiated with high-velocity ions, whereas it is normally paramagnetic. In the case of ion irradiation, the energy imparted to the target atom is large, so both Fe and Rh atoms are ejected. In contrast, in electron irradiation, the energy imparted is close to the ejection energy, so we can control the energy of the electrons. Therefore, we used electron beam irradiation to study the difference in the defect structure depending on whether only Fe or both Fe and Rh atoms are ejected.

Fe-50%Rh was irradiated with 0.8 MeV and 2.0 MeV electrons, and the defect structure was studied by means of a positron annihilation technique. The experimental result shows that the electronic structure around irradiation-induced vacancy-type defects depends on the energy of electrons.

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Study of Mass-transport Process in Noncrystalline Films Using Ion Beam Technique

H. Naramoto a), K. Narumi a), S. Sakai a), Y. Maeda a, b), T. Motooka c), Y. Ikoma c), S. Munetoh c) and X-q. Cheng c)

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The energetic ions have been employed for the position-sensitive materials processing and also for the depth-sensitive analysis of elements and defects 1). These features are typically suitable for the study of atomistic mass transport process in the near surface region, which can not be complemented by other techniques.

The mass-transport processes in crystalline solids are mostly initiated through the thermal activation of the relevant atoms bound to the surrounding atoms with the periodic nature. The recent progress in nanoscience/nanotechnology has opened the new interests in the transport process in the noncrystalline and/or the inhomogeneous atomistic environment2).

In the present report, two kinds of mass-transport related studies are described on the noncrystalline layer of Ag(40nm)/C60(100nm)/Co4C60(40nm)/C60(100nm) on MgO(100) substrate, and on ion-irradiation-amorphized Si.

The noncrystalline layers were prepared with a conventional evaporation technique using Knudsen cells by keeping the exact stoichiometry of Co4C60 mixture because the Co-C60 mixtures can be easily phase-separated above the stoichiometric ratio3, 4). The Ag top layer was deposited to avoid the influence of oxygen attack on the transport of Co atoms in the Co4C60 mixture. The mass-transport features were characterized in-situ by Rutherford Backscattering Spectrometry (RBS) equipped with a heating stage under the ultra high vacuum.

The amorphization of Si was induced by 200keV Ar ion irradiation up to the appreciable dose of 2×1016/cm2 to RBS. The additional irradiation with 10keV Ar+ ions was performed to introduce the possible bending strain in the near surface region by changing the doses systematically.

Figure 1 illustrates the typical profile changes of the layered films characterized by 2MeV 4He+ RBS. After annealing at 397K, there induced the considerable profile changes at the Ag top layer and also at the Co4C60 mixture layer. The temperature range observed coincides with that in which the compound is relaxed into more stable form as evidenced by the detailed Raman analysis14). Further annealing at 483K induced the definite change of Co profile moving toward deeper. This result is totally different from that caused by the thermal diffusion with the broadening nature towards both sides. The inward change of Co atoms could be associated with the peculiar nano-structure together with the possible bending strains.

The effect of strain on the implanted Ar distribution in amorphized Si was examined by implanting the 10keV Ar ions additionally. In this case, the compressive strains can be induced in the amorphized region below the surface.

The results obtained are not shown here for the simplicity, but the conclusions are listed below.

* The transportation of implanted 200keV Ar atoms toward the surface region was observed clearly at the higher dose of 10keV Ar ions more than 5×1016/cm2.
* The influence seems to be localized in the shallower region than the peak depth of implanted profile.
* The redistribution of implanted Ar atoms does not induce the crystallization of amorphized region.
* The systematic analysis on the observed phenomena is in progress by changing the strain parameters and the implanted species.

References
4-24 Carbon-concentration Analysis of Si Surfaces Bombarded with 10- to 100-keV C\textsubscript{60} Ions

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Our previous study shows that sputtering is one of the most important radiation effects of 10-to-100-keV C\textsubscript{60} ions\textsuperscript{1-4). It is so strong that it can affect any other radiation effect of C\textsubscript{60} ions. Carbon deposition or implantation by C\textsubscript{60} irradiation is one of such effects. Carbon content in the surface layer is sensitive to the sputtering; thus, analysis of carbon concentration could be helpful to understand the strong sputtering effect of C\textsubscript{60} irradiation. We will report here the analysis of carbon concentration of Si surfaces bombarded with 10-, 50- and 400-keV C\textsubscript{60} ions.

A piece cut from a Si(100) wafer was irradiated with C\textsubscript{60} ions. Before the irradiation, the samples were cleaned with a wet chemical treatment to reduce carbon contaminants on the surface. A part of the sample was covered with a mask, so that one sample had both irradiated and unirradiated areas. After the C\textsubscript{60} bombardment, carbon concentration in the surface layer was evaluated with a nuclear reaction \textsuperscript{12}C(d, p)\textsuperscript{13}C using 1.2-MeV D. Examples of the observed energy spectra are shown in Fig. 1. Signals at lower energies correspond to the primary D scattered from Si nuclei, while those at higher energies are attributed to protons produced in the nuclear reaction. Integrated proton yields reflect overall carbon content in the surface layer. Since the proton signals in the spectrum for an unirradiated area originates from residual carbon contaminants on the sample, the difference between the proton yields of bombarded and unirradiated areas is caused by the C\textsubscript{60}-ion bombardment.

Figure 2 shows the fluence dependence of the carbon concentration in the surface layer of C\textsubscript{60}-bombarded Si. The number of \textsuperscript{12}C nuclei per cm\textsuperscript{2} was evaluated with the use of the cross section measured by Kashy et al.\textsuperscript{5). A solid line in the figure shows carbon concentration estimated from the ion fluence. The experimental results are inconsistent with this line: At lower fluence, measured carbon concentration is higher than the value estimated from the ion fluence, while at higher fluence the increase rate of carbon concentration decreases. The former could be attributed to carbon implantation by C\textsubscript{60}-bombardment-induced recoil of the surface contaminants. On the other hand, the latter reflects the strong sputtering effect of the C\textsubscript{60} bombardment. This conclusion can be explained by the following scenario: The carbon concentration in the surface layer is determined by the competition between the sputtering and the carbon deposition or implantation by the C\textsubscript{60} bombardment. Under this competition, the higher the carbon concentration, the more carbon atoms are sputtered; thus, the increase rate of the concentration decreases with the ion fluence. It could be expected that the carbon concentration reach a certain value determined by the sputtering yield.

References

![Fig. 1 Example of energy spectra measured when 1.2-MeV D was incident on a Si sample bombarded with 50-keV C\textsubscript{60} ions. Energy spectrum measured on an unirradiated part of the sample is also shown.](image1.png)

![Fig. 2 Fluence dependence of the carbon concentration at Si surfaces bombarded with 10-, 50- and 400-keV C\textsubscript{60} ions. Solid line shows areal density which is 60 times as much as C\textsubscript{60} ion fluence.](image2.png)
4-25 1-D Crosslinked Polymer Nanowires Prepared by Single Particle Nanofabrication Techniques

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Recently we have successfully produced 1-D nanowires by the crosslinking reactions of polymer molecules within an ion track along the particle trajectory. The present nano-scaled negative tone imaging technique (single particle nano-fabrication technique: SPNT) shows a striking contrast to the conventional nuclear track technique, providing the direct formation of nanowires based on a variety of polymeric materials with fairly controlled sizes. Effective crosslinking reactions of the polymers are the unique requirement of SPNT. Thus nanowires reflecting the properties of the target polymer materials have been successfully realized by the simple one-step procedure of SPNT, giving conducting, light-emitting, ceramic, protein, hydrogel nanowires as well as the multi-functional nanowires/nanoparticles by the combination of the polymer materials. The formation process of ultra-fine optical nano-fibers by SPNT is discussed in the present letter based on silsesquioxanes (SSQ) as the target polymer material which is optically clear and suitable to the organic-inorganic conversion reactions by subsequent sintering processes.

High energy particle beam at low fluence without overlapping ion tracks produces single-ion events in the target materials, yielding a cylinder-like nanofiber in the thin film. Fig. 1 shows the AFM images of the SSQ nanofiber observed on Si substrates. The nanowires were clearly produced by all high energy particles used, and the thickness of the cross-section depends strongly on the parameters of the incident particles. The relationship between radial distribution of deposited energy released by an incident particle and induced crosslinks of polymer molecules has been precisely investigated in the previous studies, resulting in the changes of the radius of cross section of the cylindrical nanofiber. The radial distribution was characterized by the linear energy transfer (LET) of the incident particle which reflects the averaged energy density per unit length in a particle trajectory. The initial radial distribution of the released energy and the subsequent radial diffusion of the energy and/or reactive intermediates determine the cylinder-like area where gelation occurs, thus the size increased considerably with an increase in LET of the particles. Radii of the cross-section of the nanofibers were measured as 10.1, 8.7, and 6.3 nm, respectively under irradiation of 450 MeV Xe, 400 MeV Kr, and 388 MeV Ni ion beams. The values of LET of these particles are calculated as 4800, 2700, and 1800 eV/nm, respectively in SSQ thin films, which shows a striking coincidence with the values of radii. In spite of the relatively small molecular weight of SSQ (Mw = 1.2 ×10^3), the thicker nanofibers in comparison with the size observed for the other polymeric materials were observed. It has been already revealed that the radial size of the cylinder-like area depends also on the efficiency of the crosslinking reaction (G(x)). The vinyl groups in the side chain promote efficiently the crosslinking reactions, and this is the case giving considerable increase in the radius of nanofiber based on SSQ. The nanofibers based on the other silsesquioxanes were also successfully prepared by the same scheme. Sintering of the nanofibers at 1000 K caused no change in the number density of the nanofibers based on all the polymeric materials. The shrinking of the sizes was observed for the nanofibers giving 30-40 % reduction in the radii, however there was almost no change in the length of the nanofibers. The interaction at the interface restricts the axial shrinking, hence axial shrinking dominates in the present case. The successful organic-inorganic conversion by this sintering process provides the ultrafine SiO₂ optical nanofibers as demonstrated in the present study.

References
Carbon materials are well known for their excellent properties and many applications are being under intense study. The plasma enhanced chemical vapor deposition (PECVD) using hydrocarbon gas is the most used method to synthesize carbon materials. But hydrogen will be involved during processing and they will influence properties of carbon materials. In order to investigate the influences, we examine the distribution of hydrogen at near surface region in some carbon materials by a nuclear reaction analysis (NRA). We have checked the system by using a hydrogen-implanted silicon wafer. We could obtain a profile of the implanted hydrogen which was consistent with one calculated with the TRIM code. It was found that the amount of hydrogen in poly-type diamonds was 0.2-1 at %. However, it was difficult to determine the amount of hydrogen in single crystal diamonds due to a poor S/N ratio of the analyzing system.

Fig.1 Hydrogen profiles measured by the NRA. The samples were a silicon wafer implanted with 2-keV H ions/cm² (■), a polycrystalline diamond (▲) and a single-crystalline diamond (○).

References
Energetic ion bombardments can induce various kinds of effects on inorganic crystalline materials through the electronic excitations and nuclear collisions depending on the incident ion energies. The electronic excitations along a single ion passage can generate a heat pulse which results in the shear stress extending to the surface. The nuclear collisions can be influential through the generation of point defects and the associated directional flow. This process can be also employed to change the shape of the relevant samples.

In the present study, the main concern has been focused on the structural change in silicon. For the possible applications for nanopore devices, silicon-on-insulator (SOI) samples with μm-size holes made by focused-ion-beam (FIB) irradiation were prepared for the irradiation experiment. Through this experiment, the irradiation chamber was kept to be free from the carbon contamination by cooled shrouds surrounding the samples. Ar⁺ and Si⁺ ions were chosen to avoid the chemical influences on the irradiation effects. The incident ion energies are high enough to generate the displaced atoms.

Figures 1 and 2 show the transmission electron microscopy (TEM) photographs obtained from the SOI samples before and after the ion irradiation. The structural changes induced by ion bombardments were analyzed based on the dark field (DF) and bright field (BF) images combined with the selected area diffraction (SAD) patterns. The shape of each hole was determined by using the BF image, while the Si crystallinity of the hole region was judged by the DF image using the Si(004) diffraction spot and by using the SAD patterns with the resolution of approximately 500 nm. Main results are summarized as follows:

1) In the case of the irregular shaped hole (see Fig. 1), Si looks to be damaged before ion irradiation probably due to the FIB process, and no remarkable ion-bombardment effects can be observed.
2) In the case of the round hole (see Fig. 2), the hole region is crystalline before ion irradiation, but after ion irradiation the hole region is changed to amorphous resulting in an approximately 10% reduction of the hole size.
3) The observed shrinking effect may be attributed to dilation of crystalline Si upon amorphization induced by ion beam bombardments.

Further studies on the ion-beam-induced shrinking effects of the holes as well as a new method to form nanopores on SOI are currently underway.

References
4-28 Ion-induced Self-organized Ripple Patterns on Graphite and Diamond Surfaces

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The irradiation with heavy ions leads to various morphological evolutions of material surfaces as a result of a balance between roughening and smoothing processes. It is known that self-organized ripple patterns are formed on the surfaces in the case of off-normal incidence of ions. We concentrate on the study of ripple formation on surfaces of carbon allotropes after irradiation under identical conditions. According to the linear erosion theory developed by Bradley and Harper1), similar features should be observed for all different carbon materials since all surfaces become amorphous already after low fluence irradiation. Very recently, Takahiro et al.2) experimentally showed that different features appeared on graphite, diamond and amorphous carbon surfaces. In the present work, highly oriented pyrolytic graphite (HOPG) and single crystalline diamond were irradiated under identical conditions in order to investigate the allotropic effect on ripple pattern formation.

The HOPG (0001) sample (1×1×0.1 cm³ in size), manufactured by Molecular Device Tools for Nano Technology, Russia, was cleaved with a Scotch tape just before irradiation. The diamond (001) sample (0.3×0.3×0.05 cm³) is a commercially available Ib-type diamond. The samples were irradiated with Xe⁺ ions of 5–30 keV at an incident angle of 60° with respect to the sample normal at room temperature. The ion current density and ion fluence were 2–4 μA·cm⁻² and 2×10¹⁷ Xe⁺·cm⁻², respectively. The surface topography of the eroded sample surfaces was examined with an atomic force microscope (AFM) in a contact mode. Raman spectroscopy was performed by backscattering from the sample using an Ar laser operating at 514.5 nm.

Figures 1 (a) and (b) show AFM images taken for HOPG and diamond irradiated with 30-keV Xe⁺. Ripples are clearly seen on the irradiated HOPG and diamond surfaces, and they are aligned perpendicular to the ion-beam projection. The averaged ripple spacing for HOPG is found to be approximately 170 nm, much larger than that for diamond (100 nm). The line profile analysis reveals that the amplitudes of ripples, defined as height differences between hills and valleys, are typically 60 nm and 30 nm for HOPG and diamond, respectively. Thus, large differences in ripple wavelength and amplitude between HOPG and diamond were recognized. The top surface layers of Xe⁺-irradiated HOPG become amorphous as shown in Fig. 2. The SRIM simulation predicts that the crystalline diamond surface is transformed into amorphous due to sputter erosion. Further experiments under various irradiation conditions will reveal more detail in the mechanism for rippling of carbon materials. Such experiments are now in progress.

References

![Fig. 1 AFM images (upper panels) of (a) HOPG and (b) diamond irradiated with 30-keV Xe⁺ ions at 60° to 2×10¹⁷ cm⁻². Cross sectional line profiles (lower panels) are also shown. Arrows indicate the projection of ion-beam direction.](image)

![Fig. 2 Raman spectra of HOPG before (bottom) and after irradiation with 30-keV (top) and 10-keV (intermediate) Xe⁺ ions to a fluence of 2×10¹⁷ cm⁻² at an incident angle of 60° with respect to the sample normal.](image)
Optical Property Modifications of Diamond and Sapphire by Ion Implantation and Heat Treatment

Jae-Won Park a), Hyung-jin Kim a), Dong-Hwa Oh b) and Young-Chool Kim c)

a) Korea Atomic Energy Research Institute, b) Sung Kyun Kwan University, c) Hanmi G. I. Laboratory

Introduction

In order to modify the optical properties of sapphire and diamond materials, a method of the ion-implantation and subsequent annealing was employed. To understand the change in optical properties, calculation of the band structure using density functional theory (DFT) and characterizations of surface chemical properties before and after the ion implantation and subsequent annealing were conducted.

Experimental

100 keV Co and B ions using 400 keV ion accelerator and 1 MeV H ions using 3 MV Tandem accelerator were implanted into the sapphire and diamond samples with a dose of about $10^{17}$ ions/cm$^2$ for all samples, followed by post-irradiation annealing at 600 °C – 1200 °C both in air and in vacuum atmosphere for 2 – 24 hours. Once certain modifications in the optical properties of the treated diamond and sapphire specimens were visually observed, the relevant samples were analyzed with X-ray photoelectron spectroscopy (XPS) and FTIR to understand the mechanisms of the modifications. In order to examine the band gap variation in the doped diamond and sapphire, calculations by first-principles density-functional theory (DFT) were done 1).

Results and discussion

The N-ion-implanted diamond showed only black even after post-implantation annealing, but the diamond implanted with proton ($H^+$ ion) and subsequent annealing at 600 °C in vacuum emitted the purple-red color.

Since the DFT calculation shows a significant difference in the band gap between N and H doped diamonds (Fig. 1), the different elemental doping should play a major role in the optical property change. The transparent sapphire implanted with Co ions shows sky-blue colors after post-implantation annealing at 900 °C in air for 6 hours. XPS analysis shows that Co2p peak at 778.7 eV 2) which denotes metallic Co is clearly seen along with Co-Al-O peak at 782.2 eV 2) in the as-implanted sapphire (Fig. 2a) while the peak corresponding to metallic Co disappeared in the post-implantation annealed sample (Fig. 2b). Therefore, it can be concluded that the formation of Al-Co-O compound is responsible for the sky-blue color emission. On the other hand, the sapphire implanted with H, B, N showed nothing but black or white even after annealing at 900 °C.

FTIR spectra for the sky-blue colored sapphire implanted with Co ions followed by annealing show broad absorption band in the range of 588 nm to 626 nm wavelength which seems to be affected by Co ion implantation.

References

4-30 Change in Curie Temperature in Fe-Ni and Fe-Ni-Mn Alloy Thin Films Irradiated with Energetic Ions

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We have studied effects of high-energy heavy-ion irradiation on the magnetic properties in Fe-Ni invar alloys, and have already found that $T_C$ increases by the irradiation\cite{1,2}. This result suggests that the local ferromagnetic regions can be induced by the irradiation in the paramagnetic phase above $T_C$ for the matrix of Fe-Ni invar alloy. In general, high-energy heavy-ion irradiation of materials induces electronic excitation continuously along with the ion path. The induced ferromagnetic region in the irradiated Fe-Ni alloy, therefore, may have linear shape. However, it still remains uncertain whether the origin of the effect is electronic excitation or elastic displacement, because in our previous study\cite{1,2} we used bulk specimen in which the energy of the incident ions decreased with the depth. In the present work, we investigate the change in $T_C$ in Fe-Ni\cite{3} and Fe-Ni-Mn alloy thin films irradiated with several kinds of energetic ions and the dominant irradiation parameter for the modification.

Specimens of Fe-Ni and Fe-Ni-Mn alloy thin films (~500 nm in thickness) were prepared on MgO(100) single crystal substrates by means of rf magnetron sputtering at ambient temperature with target materials of Fe-32at.%Ni and Fe-28at.%Ni-7at.%Mn alloys, respectively. The specimens were annealed at 1273 K for 1 h in a vacuum below ~3.5×10^{-4} Pa. After this thermal treatment, the crystal structure of the Fe-Ni specimen changed from bcc to fcc, which was the same structure as the bulk material of Fe-Ni invar alloy. The specimens were irradiated at room temperature with 1.0-MeV $^1$H, 1.5-MeV $^4$He, 5.0-MeV $^{12}$C and 12.0-MeV $^{35}$Cl ions by using the 3-MV tandem accelerator of TIARA at JAEA-Takasaki, and 200-MeV $^{197}$Au ions by using the 20-MV tandem accelerator at JAEA-Tokai. In order to detect the irradiation effect precisely, a half area of the specimen was unirradiated by means of a masking technique, and thus both irradiated and unirradiated regions were produced in the same specimen. After the irradiation, measurements of AC-susceptibility of the specimen were performed in the temperature range from room temperature to ~550 K. The value of $T_C$ for the specimen was derived from the temperature dependence of the AC-susceptibility.

Although a large increase in $T_C$ with increasing the ion fluence was observed for $^1$H, $^4$He, $^{12}$C, and $^{35}$Cl ion irradiations, no change in $T_C$ was detected in the case of 200-MeV $^{197}$Au ion irradiation. This result suggests that the dominant energy transfer process for the increase in $T_C$ is the elastic displacement. In Fig. 1, the values of $\Delta T_C$ for all irradiations are plotted against the total energy deposited by the elastic displacement, $\Phi \times S_n$, where $\Phi$ is the incident ion fluence, and $S_n$ the nuclear stopping power for the incident ion. This result implies that the density of the elastic energy deposition is a dominant irradiation parameter for the increase in $T_C$ in Fe-Ni alloy. In the case of Fe-Ni-Mn alloy, the modification seems to be more susceptible to the elastic energy deposition than that for Fe-Ni alloy.

References


![Change in Curie temperature, $\Delta T_C$, plotted against total energy deposited by elastic displacement, $\Phi \times S_n$, for (a) Fe-Ni alloy, and (b) Fe-Ni-Mn alloy. Dotted lines are guides for the eye.](image-url)
4-31 Effects of Multi-ion Beam Irradiation on Microstructural Changes in Li$_2$TiO$_3$

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Li$_2$TiO$_3$ is regarded as one of the most suitable candidates for the solid tritium breeder material of D-T fusion reactors. Simulation of the fusion reactor environment and hence the study of a synergistic effect of atomic displacement damage in Li$_2$TiO$_3$ are presumed to be approached by a simultaneous irradiation with "triple" ion beams which consist of O$^{2+}$, He$^+$ and H$^+$ ion beams. In this study, the results of the FT-IR photoacoustic spectroscopy (PAS) with Li$_2$TiO$_3$ samples irradiated with the single ion beam, with the triple ion beams simultaneously and with the triple ion beams sequentially were analyzed for investigation of microstructural changes.

The characteristics of the Li$_2$TiO$_3$ samples and the experimental parameters used in this experiment were described in ref$^1$. The characteristic peaks in FT-IR PAS spectra for non-irradiated Li$_2$TiO$_3$ sample were observed around 680, 780, 880, 1090, 1430, 1480, 1570, 3150 and 3450-cm$^{-1}$. In comparison to the reported spectra of TiO$_2$ and Li$_2$TiO$_3$, 680- and 780-cm$^{-1}$ peaks are identified from Ti-O bond, 1570-, 3150- and 3450-cm$^{-1}$ peaks are from the O-H bond in hydroxyls adsorbed on or near the surface, and others are from C=O bond in Li$_2$CO$_3$ formed on or near the surface by reaction with CO$_2$ in the atmosphere$^1$.

Among the observed peaks, irradiation effect is clearly found for 780-, 1090- and 3450-cm$^{-1}$ peaks. In the previous study, the radiation-dose dependence of the areas of these peaks indicates that TiO$_2$, hydroxyl and Li$_2$CO$_3$ are formed near the surface by irradiation, and the amount of each was found to be strongly affected by displacement per atom (dpa). Among them, the amount of the formed hydroxyl is affected not only by dpa but also by the method of irradiation, that is, the single, the simultaneous triple or the sequential triple irradiation. This may suggest that H$^+$ implantation after irradiation degrades the generation of hydroxyl (-OH) group near the surface by trapping hydrogen in the irradiation defects or radiolytic products$^{2,3}$.

To verify the effect of H$^+$ implantation on the generation of -OH group, the results of irradiation with O$^{2+}$ ions (about 8 dpa) or He$^+$ ions (about 1 dpa) with simultaneous or without H$^+$ implantation were compared.

The results so far are summarized in Table 1. The results should be interpreted cautiously, since they are by no means complete. However, it likely supports the previous suggestion that H$^+$ implantation, especially in the sequential irradiation, suppresses the –OH group generation near the surface.

Under the fusion reactor operation, these microstructural changes may strongly affect the tritium release behavior from Li$_2$TiO$_3$, durability of Li$_2$TiO$_3$, and compatibility of Li$_2$TiO$_3$ with other components of the breeder blanket such as structural materials. Further study of the irradiation effects on these microstructural changes is needed to enhance the reliability of the related materials.

References

Table 1. Summary of the effects of H$^+$ implantation on the 3450- cm$^{-1}$ peak, which is due to the O-H bond. Notation as "A+B" means simultaneous irradiation of the beams A and B, and "A->B" means that irradiation with the beam B was performed after closing of irradiation with the beam A.

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<th>O$^{2+}$</th>
<th>O$^{2+}$ + H$^+$</th>
<th>O$^{2+}$-&gt; H$^+$</th>
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<td>(not measured)</td>
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<td>362</td>
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<table>
<thead>
<tr>
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<th>Irradiated Ions</th>
<th>He$^+$</th>
<th>He$^+$ + H$^+$</th>
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<td>≥1</td>
<td>380</td>
<td>387</td>
<td>276</td>
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</table>

Table 1. Summary of the effects of H$^+$ implantation on the 3450- cm$^{-1}$ peak, which is due to the O-H bond. Notation as "A+B" means simultaneous irradiation of the beams A and B, and "A->B" means that irradiation with the beam B was performed after closing of irradiation with the beam A.
4-32 Structural Phase Transition of One-dimensional in Wire on Si Surface Studied by Reflection High-energy Positron Diffraction

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When one monolayer of In atoms is deposited on a Si(111)-7×7 surface, In/Si(111)-4×1 super-structure is formed (see Fig. 1). From various studies, this superstructure is considered to be a quasi-one-dimensional metallic chain, which is composed of the upper and lower In atoms having two different heights. The 4×1 structure is transformed into the 8×2 structure below 130 K, accompanying the metal-insulator transition. This transition is considered to be a Peierls transition with the formation of charge-density wave from angle-resolved photoelectron spectroscopy. However, the mechanism of the phase transition between the 4×1 and 8×2 structures is not fully clarified because the atomic configuration of the 8×2 phase is still unknown. In this study, on the basis of the rocking curve analyses using reflection high-energy positron diffraction (RHEPD), we will report on the atomic displacement of the quasi-one-dimensional metallic chains due to the phase transition.

The RHEPD experiments were carried out in a UHV chamber equipped with a 22Na positron source and electro magnetic lenses. The positron energy was set at 10 keV. Two azimuthal angles were chosen to measure the rocking curves; (1) 7.5°-off oriented from the [112] direction (one-beam condition) and (2) the [110] direction (chain-parallel). The rocking curves were obtained by rotating the sample from 0.3° to 6.0° with an interval of 0.1°.

We measured the rocking curves at 210 K (4×1 phase) and 60 K (8×2 phase) under the one-beam condition, where the curve mainly depends on the vertical position of the surface atoms. The profiles of the rocking curves do not change with temperature (60 K and 210 K). To determine the vertical positions of the In atoms, we calculated the curves based on the dynamical diffraction theory. Two different vertical positions of the In atoms (d1 and d2 in Fig. 1) were used as a parameter on the basis of the structure model determined by SXRD. The measured rocking curves are in good agreement with the optimized curves at both temperatures. The vertical distances of the In atoms were determined to be d1 = 0.98±0.09 Å and d2 = 0.54±0.09 Å at 210 K, and d1 = 0.99±0.09 Å and d2 = 0.55±0.10 Å at 60 K. We found that the vertical positions of the In atoms do not change during the phase transition.

Figure 2 displays the rocking curves measured for the In/Si(111) surface at 53 K (8×2 phase) and at room temperature (4×1 phase) at the [110] direction. The rocking curves change with temperature. We calculated the rocking curves at the [110] direction using the zigzag chain structure determined by SXRD. The measured rocking curve at room temperature is in good agreement with the curve obtained by calculation, as shown in Fig. 2. On the other hand, the feature of the experimental results at low temperature cannot be reproduced by the calculation using the zigzag chain structure and even using the trimer structure. We found that the rocking curve calculated using the hexagon structure, which was recently proposed by the theoretical calculations, is in good agreement with the experiment as shown in Fig. 2. In conclusion, the metal-insulator transition of the In/Si(111) surface at 130 K occurs with the displacements of the In atoms from the zigzag chain to the hexagon structures.

References


Fig. 1. Top and side views for In/Si(111)-4×1 surface. The large black circles represent the topmost In atoms. Gray and white circles indicate the Si atoms in the first and the deeper layers, respectively.

Fig. 2. RHEPD rocking curves for the In/Si(111) surface at the [110] direction at 53 K and RT. The circles and the solid lines show the measured and calculated curves, respectively.
Reflection High-energy Positron Diffraction Study on
Surface Structures and Dynamics

Y. Fukaya a), M. Hashimoto a), A. Kawasuso a) and A. Ichimiya a,b)

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The surface atoms reconstruct to reduce the surface energy and hence the super-structures, which are different from the bulk, are formed on the surface. To investigate the surface structure and properties in detail, the surface-sensitive tool is needed. Thus, we tried to apply positron diffraction to the surface study 1). The positrons are the antimatter of electrons and have a positive charge. When the positrons are incident on the surface at grazing angle, the total reflection takes place due to the presence of positive barriers of crystals 2). The diffracted positrons under the total reflection condition are very sensitive to the topmost surface. We studied the surface structure and the dynamics of super-structures fabricated on the crystal surface, by using the reflection high-energy positron diffraction (RHEPD).

The experiments were carried out in an ultra-high vacuum chamber equipped with a 22Na positron source and electromagnetic lenses 3). The intensity of the diffraction patterns were enhanced using a micro-channel plate with a phosphor plane and recorded by a CCD camera. The accelerating voltage of positron beam was set at 10 kV. To measure the rocking curve, the glancing angle was changed from 0.5° to 6° by rotating the sample.

By depositing one monolayer of In atoms on the Si(111) surface at 673 K, one-dimensional structure of 4×1-In is formed. The 4×1 phase is transformed into 8×2 phase below 130 K. The phase transition is considered as Peierls transition. However, the phase transition is still in debate because the surface structure of the 8×2 phase is unknown. The circles in Fig. 1(a) shows the rocking curves measured from the 8×2 phase at the [1T0] direction. From the analyses based on the dynamical diffraction theory, we found that the 8×2 structure consists of hexagons, not trimers. This result supports the recent theoretical calculations. We also confirmed that the 4×1 structure corresponds to the zigzag chain model. Consequently, the 8×2-4×1 phase transition is accompanied by the large displacements of the In atoms.

When one monolayer of Ag atoms is deposited on the Si(111) surface at 773 K, the super-structure of √3×√3-Ag is formed. The phase transition from one √3×√3 to another √3×√3 phase takes place at 123 K. Although the √3×√3-Ag surface has been studied by various techniques, the phase transition remains unresolved. Figure 1(b) shows the temperature dependences of the total reflection intensities. We observed that at below 123 K, the (1/3 1/3) spot intensity drastically increases with decreasing temperature. By the intensity analyses, we found that the intensity change can be completely explained by considering the order-disorder phase transition 4).

By depositing small amounts of the Ag atoms on the √3×√3-Ag surface at 110 K, the super-structure of √21×√21-Ag phase is formed accompanied by drastic increase of the surface conductivity. Although several structure models were proposed by many researchers, there is no consensus. From the results of the rocking curve analyses, the five different structures were selected for a possible structure from candidates for the models. Finally, from the line profile of the 1/7th Laue zone (see Fig. 1(c)), we found that the additional Ag atoms are situated on the large Ag triangle of the substrate with triangular shape at the corner of the unit cell 5).

References
4-34 Characterization of Ion Beam-induced Buried Oxide Layer Using a Slow Positron Beam

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For the high-power silicon carbide (SiC) devices, SiC-On-Insulator (SiC-OI) structure is suitable. Ion implantation is one of the methods to make the SiC-OI structure. However, the properties of radiation defects have not been fully clarified. We attempted to investigate the defects in the SiC-OI structure using the positron annihilation spectroscopy which is suitable to detect vacancy-type defects.

Samples were formed by the oxygen implantation to the n-type 4H-SiC substrate with the energy of 200 keV to dose of $1 \times 10^{18}$ cm$^{-2}$. High temperature implantation was carried out for avoiding the amorphization. After implantation, samples were annealed in argon ambient at 1400 °C. The Doppler-broadening of annihilation quanta was measured as a function of incident positron energy ($E$). The obtained Doppler broaden spectra were characterized by $S$-parameters which are enhanced by the positron trapping at the vacancy defects.

Figure 1(a) shows $S$ parameters of as-implantation sample of various implantation temperatures. The mean implantation depth of $E = 4$–8 keV corresponds to the oxygen implanted region. After implantation, $S$ parameter increases in the whole energy range. This indicates that vacancy defects are formed by ion implantation. A remarkable increase of $S$ parameter in the top SiC layer ($E = 1$–3 keV) is observed. This indicates that larger vacancy clusters are created in this region. The well-defined SiO$_2$ layer is not necessarily formed because $S$ parameters of the oxygen implanted region ($E = 7$ keV, $S = 1.05$) is apparently lower than those of SiO$_2$ bulk value ($S = 1.15$). This is attributed to the existence of competitive trapping centers, such as vacancy defects which give relatively low $S$ parameters. Even if the implantation temperature increased, the clear SiO$_2$ layer was not appeared. In the top-SiC layer ($E = 1$–3 keV), a remarkable increase of $S$ parameter is observed. $S$ parameter increases with implantation temperature. This indicates that larger vacancy clusters are easily created at higher temperature.

Figure 1(b) shows the $S$ parameters obtained after annealing at 1400 °C. $S$ parameters at $E > 10$ keV decreases to the bulk value ($S = 1$) suggesting the recovery of defects. At the region of $E = 5$–10 keV, a shoulder appears. To determine the electron momentum distribution more in detail, coincidence Doppler-broadening (CDB) measurement was performed. Ratio curve of $E = 7$ keV to the SiC bulk obtained from the specimen implanted at 800 °C are shown in Fig.2. The obtained ratio curve resembles the annihilation at the interface layer of the SiO$_2$/SiC interface produced by conventional oxidation as shown on solid line in Fig.2. That is, the oxygen implanted layer contains vacancy defects with oxygen dangling bonds. In the top-SiC layer, $S$ parameters are still high even if the thermal annealing was performed. This indicates that microvoids were formed.

In summary, defects in SiC-OI structure formed by oxygen implantation were probed by positron annihilation spectroscopy. The defective SiO$_2$ layer seems to be formed in the oxygen-implanted region. Radiation defects are hardly removed even if higher implantation temperatures were applied. In the top-SiC layer, larger vacancy clusters are created. From this study, it is suggested that more advanced techniques should be developed in order to obtain a defect-free SiC-OI structure by ion implantation method.

References
Development of 2-Dimensional Nuclear Reaction Micro-analysis Technique of Boron-doped Materials

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The microanalysis of trace amount of boron has been carried out using 1 µm proton microbeam from 3MV single ended electrostatic accelerator of TIARA facility. Steel specimens containing 20-100 ppm boron have been bombarded by 1.7 MeV proton beam for gamma ray detection and 1.32 MeV H\textsubscript{2}+ beam for alpha particles. In 2006 the maps of boron distribution have been measured by detecting 3.7 MeV alpha particles from the \(^{11}\text{B} (p, \alpha \gamma) \) 8Be nuclear reaction mainly. As a result of imaging, segregation of several microns sized boron precipitates, which has been already observed by detecting gamma ray, could be clearly seen in this method.

Fig. 1. Schematic view of experimental setup for \(\alpha\)-particle measurements.

Fig. 2. A typical \(\alpha\)-particle image of 34 ppm boron contained in a steel specimen bombarded by 1.32 MeV molecular hydrogen microbeam.
Three-dimensional Measurement of Density in Minute Biological Samples by STIM

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When samples which have thickness variation and internal constitution are analyzed with an in-air micro-PIXE system\(^1\), X-ray yields have to be corrected using X-ray production cross sections and linear attenuation coefficients. For that purpose in the case of biological samples, it is necessary to measure the three-dimensional distribution of carbon, hydrogen, and oxygen, which are the matrix of cells. Because it is difficult to measure them with spatial resolution of a few microns by micro-PIXE analysis by itself, we used Scanning Transmission Ion Microscopy (STIM)\(^2\), which can measure it easily using a particle detector located just behind the sample in the atmosphere.

In our previous study\(^3\), the elemental map by micro-PIXE for a spherical sample could be corrected by STIM under the assumption the ratio of major elements in the sample was constant. Thus X-ray yields from symmetric and uniform samples can be fully corrected by STIM only. But if there are projection maps by STIM in all directions, the tomograms can be acquired in the same way as CT scan. This method is called STIM-CT\(^4\).

We attempted in-air STIM-CT of plant cells to get three-dimensional shape, and the good tomogram of the sample was acquired in 2005\(^5\). In 2006, we could get the tomogram of an insect’s head. Since it has generally hollow shape, it is necessary to measure the internal structure using STIM-CT.

In this series of study, the 3MV single-ended accelerator and the micro-PIXE system at the TIARA were used. The 3 MeV proton microbeams were scanned over a target. The diameter of the microbeam was about 1 \(\mu m\). Polycarbonate foil 5 \(\mu m\) thick was used as a beam extraction window. A target was glued on the needle just behind the extraction window, and was rotated by 180° at intervals of 15°. A surface barrier detector was placed behind the target.

Figure 1 is a photograph of the test sample. It was a head of a tiny insect. Figure 2 shows the projected image of the sample and Figure 3 is the tomogram reconstructed using the back projection method with the Shepp&Logan filter. Using all tomograms such as Fig. 3, three-dimensional reconstruction of the sample could be realized as in Fig. 4.

Fig. 1. Optical microscope image of an insect’s head, which glued on a metal needle.

Fig. 2. Projected image of the insect’s head obtained using STIM.

Fig. 3. Tomogram at line A in Fig. 2.

Fig. 4. Three-dimensional image of an insect’s head. This image was reconstructed from tomograms such as Fig. 3.

References

4-37 In-air Micro-PtXE Analysis of Asbestos in Asbestos Exposed Lung

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Asbestos were causative agents of pulmonary fibrosis and cancer. To identify inhaled asbestos in lung, lung tissues from asbestos exposed patients were analysed by in-air micro-PtXE (Particle Induced X-ray Emission). The localization of asbestos body composed by silica, iron, and magnesium were identified in the lung, and other metals of titanium and aluminium were also observed. In-air micro-PtXE was considered as useful to identify asbestos in lung tissue.

アスベストとは、天然の鉱物繊維で長さは約8 μm、幅約0.1 μm、断熱性、耐火性、防音性、耐腐食性に優れており、建築用製材として多く用いられてきた。クリソタイル（白石綿）、クロシドライト（青石綿）、アモサイト（茶石綿）などがある。アスベスト吸入は肺に炎症を惹起し、肺線維症や肺発症症と関係があるといわれている。このアスベストの肺内における局在とアスベスト組成を大気中マイクロPtXE（Particle Induced X-ray Emission）分析を用いて解析することを試みた。

対象患者: Asbestos group: Asbestos吸入歴があり、肺癌を合併した患者肺の癌以外の部分とControl group: Asbestos吸入歴がない肺癌を合併した患者肺の癌以外の部分の肺組織の凍結切片を作成し、それを表面にPtXE用の高分子フィルムを張ったスライドグラス上に載せて乾燥させた。肺組織を含むフィルム膜とともに肺切片を切り出しサンプルフォルダに固定し、3 MeVのプロトンビームを照射して、大気中マイクロPtXE分析を行った。

結果: Si, Mg, Feおよびそれらが混ざったアスベストを検出できた。得られたマイクロPtXE画像をFig. 1に示す。正常肺では組織中少量のSiを測定できたが、MgやFeはほとんど検出されなかった。一方、アスベスト肺組織中からは、無数のSiが検出されると同時に、その部位にMgやFeも一緒に検出され、無数のアスベストが肺に沈着している様子が明らかとなった。さらに、今回検出されたアスベストは、Chrysotile（白石綿, Mg3Si2O5(OH)4:わずかに鉄を含む）もしくはAmosite（茶石綿, (Mg, Fe)Si2O5(OH)2）が考えられ、Crocidolite（青石綿, Na2(Fe2+3Fe3+)8Si2O22(OH)2）ではなかった。また、アスベスト小体は組織間隙に刺さったようになっている様子が2次元的に観察できた。以上のように、大気中マイクロPtXEによる観察で、アスベストの種類や、組織内での存在部位が、明らかになった。

【考察】アスベスト暴露歴のない肺では、わずかなSiしか見られなかったが、アスベスト肺では、Siが、密度濃く、広範囲に塗りて分布している様子が見られ、明らかに多量のSiを吸入していることがわかる。さらに、アスベスト暴露歴のない肺のSiは、その部位にはほとんど金属を含まないが、アスベスト肺のSiには、Mgなどの金属が同時に存在しており、その違いは明らかであった。この成分の違いが、アスベスト患者の発症率の高さに関与するか今後の検討課題である。

References

4-38 The Present Situation and Problems of the Analysis of Boron Micro-distribution in Tumor Cells Using Micro-PIXE and PIGE

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We have applied micro particle induced X-ray emission (micro-PIXE) to determine inter- and intracellular distribution of boron (10B). Because the energy of micro-PIXE from 10B is too low, we employed a method to detect gamma-ray produced from the nuclear reaction of 10B(p, αγ)7Be, namely particle induced gamma-ray emission (PIGE). We showed the results of inter and intracellular distribution of boron elements in tumor tissue using micro-PIGE. However, we analyzed many background too and still can not detect the true boron elements. So further investigation is necessary to obtain higher spatial resolution and the optimization of the measurement time or the improvement of the sampling methods should be investigated.

In future, this method will be applied to analyze the intracellular micro-distribution of the capture element and development of new drugs for NCT.

References
4-39 Standard Reference Material for Determination of Trace Elements in Biological Materials by Micro Beam PIXE

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Standard Reference Material (SRM) is indispensable to the calibration of apparatus and the evaluation of the accuracy and precision of analytical technique. Previous work, we showed utilities of several macro porous anion and cation-exchange resins for micro PIXE analysis. In this work, we made SRM containing the known amount of Ca\(^{2+}\) and trace amount of essential elements for living matter, such as Fe\(^{3+}\), Ni\(^{2+}\) and Zn\(^{2+}\) in cation-exchange resin. Macro porous anion-exchange resin, Macro-Prep 25S (BioRad) was suspended in 0.01 M HNO\(_3\) and added the known amount of standard solutions. The concentration of trace elements in the resin was 20-60 ppmv. Individual particle of the resin was subjected to 3 MeV proton bombardments by micro beam system of TIARA. It was found the macro porous nature of the resin (pore size; 72.5nm) allows cation to access the exchange sites located throughout the matrix. This SRM is similar to biological cell because it is spherical in shape and its matrix elements are carbon, oxygen and hydrogen. This SRM made of macro porous resin has a possibility for use as SRM for determination of trace elements in biological samples by micro beam PIXE.

Fig.1 Relationship of Concentration Trace Elements and Relative X-ray Intensity of based on Ca K\(\alpha\)

References
4-40 Measurement of BrdU in Mouse Brain by In-air Micro PIXE Analysis

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1. Introduction

There are many reports that trace elements might be related to cerebral nerve diseases, such as Alzheimer’s disease1). To examine changes of spatial distributions of trace metal elements in cells, organs or tissues using various kinds of disease model, it is useful to establish a new pharmaco-therapeutics.

In this study, we developed a method to analyze elements in brain slices using histamine receptor gene knockout mice on the basis of Micro-PIXE analysis.

2. Experimental method

1) Target preparation of brain samples for PIXE analysis

We examined 4 target preparations: ① brains were removed and stored at –80°C, ② brains were removed and immediately quick-frozen in 2-methylbutane cooled in liquid nitrogen, ③ brains were removed and treated with sucrose solution and stored at –80°C, ④ brains were removed and immediately quick-frozen in dry ice to –40°C.

The target preparation of ④ was most suitable for this study. Trace metal elements were difficultly detected in the frozen conditions of ① and ③. In the condition of ②, the brains were delicate and it was difficult to make their slices.

2) Histamine receptor gene knock out mice and their neurogeneses

Histamine H1 receptor gene knock out mice (H1-KO) and their wild type mice were bred in our laboratory. 5-bromo-2’-deoxyuridine (BrdU) of 75 mg/kg were injected into mice 4 times at 2-hour intervals. Each brain was cryosectioned sagittally at 16μm thickness. The brain slices were mounted on polycarbonate films, which were processed with 5N sulfuric acid. The brain slices were examined by the micro-PIXE analysis with 3 MeV-proton micro-beams of beam currents of ~120 pA.

3. Results and discussion

H1 receptor antagonists are widely used to such as a medicine for a cold or to relieve itching. However it is well known that sedative antihistamines induce cognitive decline in human through blockage of H1 receptor. H1KO mice showed impaired leaning and memory than their wild type (C57BL/6) mice. It was recently reported that newborn cells within 24 hours in the hippocampus are related to the cognition. BrdU labeling hippocampal neurogeneses were observed in the brain slices of H1KO and their wild type mice by the use of micro-PIXE analysis. Figures 1 and 2 show the beam irradiation image of a brain slice and the distribution image of bromine in the region of 100 × 100μm² in the brain slice of Fig 1.

![Beam irradiation image](image1)

![Image of bromine of brain slice of wild type mouse.](image2)

Fig. 1 Beam irradiation image   Fig. 2 Image of bromine of brain slice of wild type mouse.   (100 × 100μm²)

![Amount of bromine in 1μm² of brain slices](image3)

Fig. 3 Amount of bromine in 1μm² of brain slices

The amounts of bromine in the slices of wild type mice and H1KO mice are shown respectively in Fig. 3. The amount of Bromine in the case of H1KO mice is small compared with that of wild type mice. This result is accord with the results of other physiological studies using H1-KO and their wild type mice2).

Here, we presented the difference of accumulation of BrdU in the brain of H1-knock out mouse from that of its wild type mouse by the use of micro-PIXE which can be used to clarify the roles of trace elements in psychotic diseases such as depression, schizophrenia and Alzheimer’s disease.

4. References

4-41 Monte-Carlo Particle Trajectory Simulation Study on Classification of Structure of 3-MeV C\(_3\) Cluster Ion

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We have studied interaction of swift cluster ion beam with matter. The interaction is well described by the nuclear collision of projectiles and target atoms with the many body effect, as far as the projectile speed is lower than the Bohr speed \(v_B = 2.19 \times 10^7\) m/s. When the speed exceeds \(v_B\), the interaction of the swift cluster ion is caused mainly by the electronic collision, and the interaction mechanism has not yet been revealed. The interaction caused through the electronic collision involving a many body effect of projectile itself suggests us necessity for revealing the relation between a projectile charge state and time interval of irradiation between each constituent ion or a cluster spatial structure. We have therefore developed a measurement system\(^1\), which realizes simultaneous observation of charge states and a planar pattern of cluster constituent ions.

The Coulomb explosion imaging technique is utilized in order to expand the space size of the 3-MeV C\(_3\)\(^+\) ion from angstrom- to millimeter-size in our system, which is consisted of the thin carbon foil and a screen of a luminous type of multi-channel plate. It was required to evaluate the accuracy of our measurement system\(^1\), which realizes simultaneous observation of charge states and a planar pattern of cluster constituent ions.

The dependence of the accuracy on the radii was also calculated and applied for the decision of the radii. We, finally, evaluate the accuracy of our measurement as 70 \(\%\) and 90 \(\%\) for triangular and linear structures respectively, when \(R_{in} = 0.67\) and \(R_{out} = 1.75\) for triangular and linear structures, respectively.

In conclusion, we developed the evaluation method for our charge state and cluster structure measurement system, based on the Monte-Carlo particle trajectory simulation. The calculation results also provide us a criterion for the structure classification, \(R\), with accuracy. The relation between the charge state and the structure, classified using these radii, qualitatively agrees with the one theoretically predicted by Kaneko\(^5\).

**References**
Study of Secondary Ion Emission from HOPG Target Bombarded by Fast Cluster Ion Beams

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Recently the interaction of fast (MeV) cluster ions with solids has attracted attention in basic physics and application fields. Fast cluster ion bombardment causes nonlinear effect or synergetic effect, which is unusual for single atom collision with solid targets. A time of flight (TOF) mass spectrometer combined with pulsed fast cluster ion beams which were produced by the TIARA tandem accelerator was used for a secondary ion measurement. In this report we present some results of positive and negative secondary ion measurement from a highly oriented pyrolytic graphite (HOPG) target bombarded with carbon, aluminum and gold cluster ions.

高速（高エネルギー）クラスターイオンと物質との相互作用では単原子イオンの数の集合体として線形的に振る舞うのではなく、個数以上あるいは個数以下の非線形的な効果を与える。二次荷電粒子放出の収量や二次イオン放出スペクトルの測定を通じて、高速クラスターイオン照射の特徴を調べることが本研究の目的である。H18年度は直線飛行時間型（TOF）質量分析器を用いて炭素、アルミニウム及び金クラスターイオンとHOPG（Highly Oriented Pyrolytic Graphite）標的との衝突で放出される正負の二次イオン測定を行ったので報告する。

同種のクラスターイオンで核子当たりのエネルギーを広範囲に変えることができることが望ましいが、加速器の制約上不可能なので、加速可能な粒子を変えることで核子当たりのエネルギーを変更し比較した。

標的試料にはHOPGを用いた。HOPG試料は层数に剥がすことができ新しい表面を測定に使えるので他の試料に比べて不純物の吸着が少ないと考えられる。それでも相当不純物（例えば、水、ナトリウムなど）が観測されるので、ECRイオン源からのArビーム照射により表面のクリーニングを行ったのち、TOFスペクトルの計測はイオンビームをパルス化し、TAC（時間波高変換）を用い方法により行われた。核子当たりのエネルギーはBohr速度（〜25 keV/u）とその前後を選び、放出正負二次イオンの全収量（水素を除く）の入射クラスターイオン照照射に対する依存性を比較した。

次にイオン放出現象においては、イオンの速度がBohr速度以下の低速領域では、核的衝突が支配的でクラスターサイズに余り依存せずほぼ一定で、負の非線形性を示している。一方、C及びAlの場合はクラスターイオン的に、ほぼ指数的に増加し、正の非線形性を示している。すべての入射クラスターについて、負イオンの収量はイオンの数倍から数10倍となっていて、この傾向はエネルギーに関わらず同様である。この理由は未だ明らかではない。

二次イオン放出実験においては、イオンの速度がBohr速度以下の低速領域では、核的衝突が支配的でクラスターイオンでは負の非線形性が、またBohr速度以上の高速領域では電子的衝突が支配的で正の非線形性が表れることが理論的に知られており1), 今回の実験では定性的には理論と同じ結果になっている。

References
4-43 Secondary Ion Emission from Polycarbonate upon Cluster Ion Impacts

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Secondary ions are ejected from the surface when the primary ions bombard the target. Cluster ion bombardments give different secondary ion emission yields from those for monoatomic ions because of their peculiar irradiation effect. We have developed a time-of-flight (TOF) secondary ion mass spectroscopy using cluster ion impacts, and applied it to various samples1-3). In this paper, we report secondary ion emission from polycarbonate (PC) upon cluster ion impacts.

Positive secondary ion (P-SI) TOF measurements for amorphous PC films with a thickness of 100 μm were performed with a linear type TOF mass analyzer using pulsed monoatomic and cluster ion beams produced by a 3 MV tandem accelerator of the TIARA1-3). A schematic diagram of the experimental arrangement is shown in Fig. 1. Pulsed 0.5 MeV/atom-C3 + and C8+ beams were injected into the sample with an incident beam angle of 45º relative to the sample surface. P-SIs accelerated between the sample and a TOF drift tube flew in the tube before hitting a 40mm-diameter microchannel plate (MCP). The TOF spectra recorded in a multi-channel analyzer (MCA) were transferred to a computer. Several improvements were made in the TOF system detailed in our previous paper1): (1) the mass resolution was improved by shortening a half-width of the pulsed incident ion beam to ca. 0.1μs and extending a length of the TOF drift tube, (2) the SI transmission efficiency of the analyzer was enhanced by inserting an electrostatic lens with transmission grids between an extractor grid and the MCP. The TOF measurements were performed at a pressure below 3 × 10^{-8} Torr and a fresh sample was used for each measurement.

We note that the relative intensities of the fragments are different between C1+ and C8+ impacts although the PC samples are bombarded with the primary ions of the same element with the same velocity. Implanted ions lose their kinetic energy by interaction with the target. Depending strongly on the local density of energy deposited by the implanted ion, the target molecule may be fragmented and a fraction of the fragments is emitted as secondary ions. The local density of the energy deposited at the near surface due to C8+ impact is larger than that due to C1+ impact because constituent atoms originating from the same cluster simultaneously transfer their kinetic energy to a small area of the target surface. The higher deposited energy density makes the fragmentation process for C8+ different from that for C1+; this difference provides the different positions and relative intensities of some fragments between the C1+ and C8+ impacts.

In conclusion, the cluster ion impact provides different relative P-SI intensities for PC from those for the impact of the monoatomic ion with the same element and velocity.

References
4-44 Amorphization of Si Single Crystals by Au Cluster-ion Irradiation

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Si single crystals were irradiated with Au cluster ions, and the amorphization of the crystal was observed by means of a transmission electron microscope. The experimental result shows that the degree of amorphization strongly depends on the size of Au cluster ions.

原子が複数個の化学結合状態を形成して電荷を帯びた、いわゆるクラスターイオンを高エネルギーに加速して物質に照射すると、単原子イオンを同じ個数だけ照射した場合と異なる効果が期待される。それは、ある領域に原子数複数個に対応するエネルギーが同時に対象されるためである。前報では、Si単結晶をAu ₂、Au ₃クラスターイオンで照射し、透過型電子顕微鏡(TEM)で欠陥集合体を観察した結果を示した。それによると、Au原子の個数を同数照射したにもかかわらず、クラスターサイズが増加するにつれて観察される欠陥集合体の数やサイズが増加することが示された。これらの結果は、クラスターイオン照射量が少ない場合の結果である。今回は、照射量が多いとき、Siにどのようなクラスター効果を現れるかを調べた。

ターゲットには111方位のSi単結晶を用いた。あらかじめTEM観察用に薄膜化した試料をTIARAタンデム加速器により、1原子あたり1.66 MeVのAuイオン、Auイオン、Auイオンを室温で照射した。クラスター効果をクリアに見るために、照射されたAu原子の個数が一定になるような照射量を用いた。TEMにより照射後の試料の電子回折パターンを観測した。

Fig.1 Electron diffraction patterns for Si single crystals irradiated with Au cluster ions (a)Au, (b)Au, (c)Au. Irradiation fluence is 2 ×10¹³Au/cm².
4-45 Double Differential Cross Section for Neutron Production of Beryllium

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For boron neutron captured therapy (BNCT), there is a candidate as a accelerator neutron source for the use of Fixed Field Alternating Gradient (FFAG) ring, in which protons with the energy of about 10 MeV are circulated. In this source, thin beryllium will be used as a target material. Neutron production yields, its energy spectra and angular distributions from the thin beryllium bombarded by protons of about 10 MeV have not been measured systematically and there is no simulation code to estimate exactly these items. In this work, neutron spectra and its angular distributions from the beryllium target bombarded by protons were measured for the characterization of neutron source intensity, source term of shielding calculation and development of simulation code including of PHITS code [1].

Measurements were performed at the No.1 heavy ion irradiation room of AVF cyclotron in TIARA. Neutron energy spectra were obtained by the time-of-flight method using the two NE213 organic liquid scintillation detectors with 5.08-cm-diameter and 5.08-cm-thickness. The thickness of beryllium target in the vacuumed chamber is 15 μm which is corresponding to the energy loss of 940 keV for 10 MeV protons. The neutrons and photons emitted from a beam dump, in which the transmitted protons through the target are stopped, are shielded by iron and paraffin blocks settled between detectors and the beam dump.

Figure 1 shows the neutron production double differential cross section for the energy above 0.8 MeV toward 15 degree direction compared with values of Evaluated Nuclear Data File, ENDF/B-VII. Peak energies and values of the cross section are the almost same between measured and evaluated ones. Two peaks in the measurements are corresponding to the energy levels of the ground state and the first excited state, 2.4 MeV level [2]. For the evaluated spectrum, there is a level corresponding to energy of 1.4 MeV, but such a peak is not appeared in observation. Similar tendency is observed in the neutron spectra of other direction angles.

Figure 2 shows the angular dependence of neutron production yields. The term of “Two peaks” presents the integrated neutron yield with the energy region above 4 MeV including two peaks, “Low energy” presents that with the energy region below 4 MeV, and “Total” presents the sum of “Two peaks” and “Low energy”. As three types of neutron production yields get sharply decreasing with backward angles above 45 degree, the irradiation position of the patients should be forward direction below 45 degree to secure the neutron intensity for radiation therapy.

References
Development of Dose Monitor Applicable to Wide-energy Neutron

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Workers in high-energy accelerator facilities, such as J-PARC, are potentially exposed to high-energy neutrons, photons and muons because of their high penetrability in radiation shielding. Measurement of doses given by these particles is therefore of great importance for assuring radiation safety in the accelerator facilities. None of the existing survey instruments is however able to measure doses given by all of these particles. The authors have developed a new radiation dose monitor, designated as DARWIN (Dose monitoring system Applicable to various Radiations with Wide energy raNgnes), for real-time monitoring of doses in workspaces and surrounding environments of high-energy accelerator facilities 1). In the fiscal year 2006, we have measured doses in various radiation fields in order to demonstrate the applicability of DARWIN.

Figure 1 depicts the time-dependence of neutron dose rates measured with DARWIN in the Light Ion Room No.3 of TIARA, together with that of the beam current of the cyclotron. Quasi-monoenergetic neutrons of 60MeV were generated by the 7Li(p, n) reaction in the LC0 course and then transported to the Light Ion Room No.3. The time profile of neutron dose rates measured by DARWIN is consistent with that of the beam current. It was confirmed that the measured dose rates agree with the estimated dose rates from the neutron spectrum in the field. These results indicate the rapid response and reliability of DARWIN.

Figure 2 shows the measured dose rates with DARWIN in comparison with the corresponding estimated values in various fields. The measurements were performed in 1) TIARA, 2) HIMAC at the National Institute of Radiological Sciences, 3) Los Alamos Neutron Science Center (LANSCE), and 4) the neutron fields of 235U and 241Am–Be sources in the Facility of Radiation Standard (FRS) at JAEA. Natural background dose rates of neutron from cosmic-rays were also measured in Tokai-mura and Los Alamos and then compared with the estimated values calculated by EXPACS 2), EXcel-based Program for calculating Atmospheric Cosmic-ray Spectrum. Excellent agreement can be found between the measured and estimated dose rates over wide dose rates.

For further investigation concerning the sensitivity of DARWIN, the background neutron dose rates in Tokai-mura were measured simultaneously with DARWIN and a conventional rem-counter (TPS-451, Aloka). It was found that DARWIN needs only 20 min to obtain the data with a statistical error within 10%, while the rem-counter needs ~ 5 h. This result clarifies that one order of magnitude for difference in the sensitivity for neutron radiation lies between DARWIN and the rem-counter. With this property, DARWIN enables us to monitor small fluctuations of neutron dose rates near the background level.

References
2) http://www.jaea.go.jp/04/nsed/ers/radiation/rpro/
3) EXPACS/expacs.html

Fig.1 Relation between the dose rates measured with DARWIN in the Light Ion Room No.3 of TIARA and the beam current of the cyclotron.

Fig.2 Measured dose rates with DARWIN in comparison with the corresponding estimated values in various radiation fields.
4-47 Development of a Detector for Absolute Measurement of Neutron Fluence in Quasi-monoenergetic Neutron Calibration Fields of High Energies

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In facilities of high intensity proton accelerator, it is important for radiation control to measure the dose for high energy neutrons penetrating the shielding materials. High energy neutron calibration fields are necessary for accurate dose measurements to appropriately calibrate the monitor and the dosimeter used there. For the neutron fields above 20 MeV, however, the international standards for the neutron fields and relevant calibration techniques have not been established. Therefore, by using the quasi-monoenergetic neutron irradiation fields of several tens of MeV at TIARA1) of Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency (JAEA), the development of the calibration fields for such high energies has been in progress toward the standard fields. Neutron fluence of peak region is one of the most important characteristics of the calibration fields with quasi-monoenergetic spectrum. For absolute measurement of the peak neutron fluence, a proton recoil telescope (PRT) has been developed.

The PRT consists of a Si semi-conductor detector to measure ionization energy loss (ΔE) and an organic liquid scintillation detector to measure full energy (E) of recoil protons. The Si detector has the sensitive area of 3000 mm² with a fully depletion layer of 300-μm. The liquid scintillator (BC-501A type) has the sensitive volume of 7.62-cm both in diameter and in length. Its performance was tested in the neutron fields, where neutrons were produced in the nuclear reaction of $^7$Li targets with 50, 65 and 80 MeV protons. The proton energies correspond to neutrons with the peak energy of 45, 60 and 75 MeV, respectively. The measurement is based on the detection of the recoil protons produced in a polyethylene converter by the incident neutrons. For the precise measurement of the peak fluence based on the n-p elastic scattering cross-section, the unwanted protons from carbon in converter material (C(n,xp)) must be subtracted. Therefore, measurement with graphite converter was also performed. The PRT was set on the neutron beam axis at the two different distances from the target (5.585 and 12.905 m). The brass shadow bar (8-cm in dia., 50-cm in length) was used to avoid the neutrons directly incident to the detector. The thin annular converters with inner diameters of 8.8-cm were applied. Since the collimated neutron irradiation fields at TIARA enlarge with increasing distance from the target2), outer diameters of the annular converters were appropriately selected to use the irradiation fields effectively.

Recoil protons were clearly identified in the obtained ΔE v.s. E distribution as shown in Figure 1. Energy resolution of the Si detector was 24.7 % for recoil protons corresponding to peak neutrons with 45 MeV, and was enough for the separation of protons from background deuterons. By selecting the recoil protons, energy spectra of the recoil protons were obtained for the two different converter materials. After the compensation for the backgrounds from carbon and air nuclei, the recoil proton energy spectra only for n-p elastic scattering were derived. From the results for the three neutron fields with 45, 60 and 75 MeV peak, it was confirmed that detection efficiency was enough for the absolute measurements of peak neutron fluence both at 5.585 and 12.905 m from the target.

References

Fig.1 Energy loss (ΔE) measured by the Si detector v.s. Energy (E) measured by the organic liquid scintillation detector for measurement with polyethylene converter (neutrons with 45 MeV peak).
Development of Focusing High-energy Heavy Ion Microbeam Technology and Its Application

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\textsuperscript{a)}Department of Advanced Radiation Technology, TARRI, JAEA, \textsuperscript{d)}21st Century COE program Graduate school of medicine, Gunma University, \textsuperscript{c)}Radiation-Applied Biology Division, QuBS, JAEA, \textsuperscript{d)}Environment and Industrial Materials Research Division, QuBS, JAEA

A focusing type high-energy heavy ion microbeam system with a sub-micron level spatial resolution had been established in the vacuum system\textsuperscript{1)}. Now an end-station was developed to extract microbeams out to the atmosphere through a thin film, so as to evaluate response of specific part of intracellular structure such as a cell nucleus, a cytoplasm or cell wall with a high spatial resolution of 1\textmu m level.

Figure 1 shows the schematic draw of an end-station, which was connected with a lower plate of the Q-lens frame through a flange of the ICF-70 standard, was simply designed to evaluate the beam size at the beam exit window on the atmosphere, and to perform the preliminary microbeam applications. It has flexibility of the bellows-seal and positioning mechanism with a movable range of \pm12.5mm in each direction by X/Y stage with stepping motor drive. The manifold connected to the bellows is 42mm in outer diameter, 32mm in inner diameter, and 49mm in length. It has two ports for a secondary electron detector; one is for a light-guide connected to a ring-type plastic scintillator set at a height of 14 mm from the under-surface of the manifold to detect scintillation by secondary electron from the sample, and the other is for high-voltage supply on a moralized surface of the scintillator. On the bottom of the manifold, there are three penetration holes with diameters of 1 mm, 2 mm and 3 mm, respectively. A copper-grid (1000 lines per inch) standard sample, which was utilized to measure the beam size by secondary electronic mapping, and a plastic scintillator film to monitor beam spot profile at the target were attached outside of the 1-mm and 3-mm holes, respectively. The 2-mm hole was a blank for use of irradiation experiments. All holes together with the grid and the film were covered with a polyimide thin film of 7.5\textmu m thickness to keep the vacuum inside of the chamber. The flat bottom of the manifold was designed, so that an air-gap between the film and sample should be as small as possible in the irradiation set-up for the experiments.

Figure 2 shows a typical secondary electron image of the grid at the beam exit window obtained with this new setup using a microbeam of 260MeV Ne ion. The average beam size was obtained at about 2\textmu m from the peak fitting for the secondary electron yield curves along lines for X and Y directions, respectively, so far. The spatial resolution of the beam at the sample in the atmosphere can be deteriorated by scattering in the film and the air gas.

Further work will focus onto reliability in forming the higher spatial resolution microbeam, which depends on stabilization of a beam current from a cyclotron, as well as on establishment of automated single ion hit system.

Reference

Development of Irradiation Position Control Techniques for Ion Microsurgery Using an Ion Beam Induced Fluorescent Analysis

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a)21st Century COE Program, Graduate School of Medicine, Gunma University, b)Department of Advanced Radiation Technology, TARRI, JAEA, c)Environment and Industrial Materials Research Division, QuBS, JAEA, d)Department of Medicine, Graduate School of Medicine, Gunma University, e)Gunma University Heavy Ion Medical Center, Gunma University,

The heavy ions have unique characteristics of high energy transfer localized in the vicinity of almost straightforward tracks. The heavy ions are suitable for the treatment of small tumors with the size of several millimeters or less. This research aims at developing an innovative microsurgery treatment system, using the carbon-ion pencil beam, for AMD (age-related macular degeneration). The AMD treatment system has been studied using the carbon-ion pencil beam system.
Development of the Cyclotron System for Advanced Irradiation


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Microbeam Formation

For production of a focused heavy-ion microbeam\(^1\) in the JAEA AVF cyclotron, the beam energy spread has been improved by optimizing the operating parameters including flat-top (FT) acceleration\(^2\). In the case of a 260 MeV-\(^{20}\text{Ne}^+\) beam, the energy spread of 0.05 % was achieved and both of the spatial resolution and the hitting accuracy were evaluated at less than 1 \(\mu\text{m}\)\(^3\). In the next place, the FT acceleration of a 220 MeV-\(^{12}\text{C}^5^+\) beam was tested. The beam phase in the FT acceleration was controlled by a pair of phase defining slits in the center region of the cyclotron. The beam current was as high as 0.6 \(\mu\text{A}\) when both of the sinusoidal and the saw-tooth waveform bunchers were turn on. As shown in Fig. 1, we observed the clear turn separation of the beam intensity distribution with the deflector probe. The extraction efficiency at the deflector was successfully increased up to 98 % with the single-turn extraction.

In order to reduce the sources that cause instability of ion beam current, we have exchanged the temperature control system of magnet power supplies. By the temperature control of the feedback control circuit with Peltier devices, a beam current fluctuation caused by power supplies was significantly reduced.

Uniform Irradiation Using Multipole Magnets

We are now developing the uniform-beam irradiation system using multipole magnets with the aim of the advance in uniform-irradiation technique. It enables us to obtain homogeneous irradiation effects on over a sample with a large area or on a large number of samples simultaneously without scanning the beam.

We have theoretically studied the beam dynamics under the nonlinear magnetic fields to produce a beam with a uniform transverse intensity profile in the beam transport at the AVF cyclotron facility\(^5\). Although all the odd-order multipole magnetic fields are, in theory, required for uniformization of a Gaussian beam, it is practically possible to produce a uniform beam only with an octupole focusing. Its integrated strength is \(1/(\epsilon\beta_0 \tan \theta)\), where \(\epsilon\) is the rms emittance of the beam, \(\beta_0\) is the beta function at the octupole magnet, and \(\theta\) is the betatron phase advance from the magnet to a target. The full width of the resultant uniform is \(2\pi \sqrt{|\epsilon_0| \cos \theta}\), where \(\epsilon_0\) is the beta function at the target. Based on these formulae, we have optimized the optics of the beam line. It is possible to produce a beam with a uniform profile of 10 cm square at the target. The effect of beam emittance and energy spread on beam uniformization has also been explored through systematic simulations.

Intensity Control with Beam Attenuators

Beam current control over a very wide range is required for experiments and beam tuning in the JAEA cyclotron. A beam attenuator (ATT) to attenuate the beam current without changing the beam profile is set in the injection line of the cyclotron. The beam current is controllable in the range of \(10^{-1}\) to \(10^{-12}\) by combining the ATT meshes of \(10^{-3}\), \(10^{-2}\) and \(10^{-1}\). It was found that the ATT did not always keep the beam profile at a target point and did not control the beam current precisely according to the combination of the meshes. We estimated the beam size at the installation point of the ATTs with the design value of the emittance. Influences of combination of the meshes to the beam profile was also examined by the Gaf film which allows the beam profile measurement easily. As a result, we realized that the intervals of the meshes were too short at the beam diagnostic station of IS2, and the spacing of the holes in the mesh was too long at that of IS5, which is a waist point in the beam transport.

References

4) Y. Yuri et al., in Proceedings of the 3rd Annual Meeting of Particle Accelerator Society of Japan and the 31st Linear Accelerator Meeting in Japan, 913 (2006)
4-51 Development of All-permanent-magnet ECR Ion Source for the JAEA Cyclotron (Ⅱ)

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We have been developing an all-permanent-magnet type ECR ion source (LECR) since 2003 to realize the high stability of beam intensity and reduction of the adjustment time of the JAEA AVF cyclotron\(^1\). The magnetic field distribution of LECR is changeable by an axially moving the mirror tuning magnets of the extraction and the gas injection side\(^2\). The beam current of LECR increased 10 times as much as that of the original design, and the continuous operation became possible by the previous modification\(^3\). In this paper, we report the new modification for further increase of beam intensity and the result.

In the original design, the entire magnets of LECR can be slide in the axial direction within 60 mm to adjust the relative position of the beam extraction hole against the magnetic field. The vacuum was deteriorated by outgas from the inner wall of long puller by hit of extracted beam. It appeared that the distance of 5 mm in the axial direction is enough by experiment. Therefore, we modified design of the high-voltage insulation and the joint parts of the plasma chamber, and changed the diameter of einzel lens.

There was a clearance of 2 mm between the plasma chamber and the magnet for the high-voltage insulation sheet. The inner diameter of the plasma chamber was extended to increase confinement efficiency by decrease in clearance of 2 mm. The material of a plasma chamber was changed into oxygen free copper from stainless steel to improve the cooling efficiency.

A schematic drawing that describes the modification is shown in Fig 1, and the modification is summarized in Table.1.

The improvement of LECR by the modifications is shown in table 2. The beam current of O\(^{7+}\) increased from 0.6 to 1.8 µA. A considerable amount of beam loss may takes place in the analyzing magnet because its chamber inner gap is 54 mm, not enough for beams from an ECR ion source with a large emittance.

We are, therefore, planning to install LECR in an injection line of the cyclotron with an analyzing magnet whose inner gap of the chamber is designed 92 mm.

References

Table.1 the modification in FY2006

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<td>44 33 18 1.8</td>
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<tr>
<td>Materials of plasma chamber</td>
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<tr>
<td>Puller length</td>
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<tr>
<td>Diameter of einzel lens</td>
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Table.2 The performance of LECR

<table>
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<th></th>
<th>O(^{4+})</th>
<th>O(^{5+})</th>
<th>O(^{6+})</th>
<th>O(^{7+})</th>
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<tr>
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<tr>
<td>LECR (2005)</td>
<td>16 15 10 0.6</td>
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<td>LECR (2006)</td>
<td>44 33 18 1.8</td>
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Fig.1 The modification of LECR ion source in 2006.
4-52 Development of Beam Generation and Irradiation Technology for Electrostatic Accelerator


Department of Advanced Radiation Technology, TARRI, JAEA

1. Measurement of beam-energy spread in a 3 MeV TANDEM accelerator

The monochromatism of the beam energy is the most important for the highly precise irradiation such as a micro-beam technique, and it great depends on the stability of the terminal voltage in the electrostatic accelerator. We aim at establishing the accurate measurement system of the beam-energy spread to evaluate the acceleration performance. In the TANDEM accelerator, the monochromatism of the beam energy would be affected from not only the terminal voltage stability but also from the energy loss by the collision with the N₂ stripper gas. Therefore we tried to measure the beam-energy spread dependent on the stripper gas pressure by using the nuclear reaction with narrow resonance width (150eV) in 

\[ ^{24}Mg(p, \gamma)^{25}Al \]

The result of the Fig.1 suggested that this measurement system could accurately evaluate the monochromatism of the beam in 10⁻⁵ order.

Fig.1. The relation between the beam-energy spread and the stripper gas flow. The horizontal axis is the vacuum in an acceleration tube relative to the stripper gas flow.

2. Voltage calibration for TIARA 3 MV single-ended accelerator using a resonance of RBS

The voltage calibration has been carried out using the resonance nuclear reactions of 

\[ ^{27}Al(p, \gamma)^{28}Si \] and \[ ^{24}Mg(p, \gamma)^{25}Al \] with narrow reaction energy spreads. The incident proton beam energies for the former and the latter are used at 992 keV, 1317 keV and 2010 keV, 2400 keV, respectively. The calibration at 3 MV was difficult because a resonance reaction with a narrow energy spread did not exist in the vicinity of the incident proton beam at 3 MeV. Therefore the resonance for the Rutherford Back Scattering (RBS) of \( ^{16}O, ^{3}He \) at 3.04 MeV at a direction of 180 degrees was used for the calibration of the accelerator voltage. The thin TiO₂ layer of 50 nm was evaporated on glassy carbon as a sample. The sample was placed on the 15 degree beam line (SC). The He⁺ beams generated by the accelerator were injected into the sample. The back-scattered He⁺ particles were detected by a SSD detector at 170 degrees. The He⁺ yields were measured as a function of the He⁺ energy.

The difference between the indicated voltage of the accelerator controller and measured one, 3.04 MV, was estimated to be 1%.

3. Acceleration of Ge cluster ion to MeV energy region

We tried to accelerate Ge cluster \( (Ge_n : n=2-4) \) ions up to 6 MeV with a tandem accelerator for applying them to the study of interaction between swift cluster ions and matter. Negative Ge cluster ions are generated by a Cs sputter ion source. The mass spectrum of negative ions extracted from the source is shown in the Fig. 2. We can recognize Ge clusters \( (Ge_n) \) up to Ge₂ in the beam current of nano ampere order. Clusters of Al are also obtained because Ge-Al alloy is used for the sputter cathode to stabilize the beam current. Ge cluster ions, which have the medium mass between that of a carbon cluster and that of a gold cluster, are required by users.

Fig.2. The mass spectrum of cluster ions extracted from the ion source

4. Study of Faraday cup (FC) for a fullerene ion beam current measurement

A current of fullerene beam was difficult to measured with a conventional FC, because a large number of secondary positive ions produced by fullerene collision escape through the electron-suppression field. Therefore we developed of the FC that applied the external magnetic field (430 Gauss) to a conventional FC for suppressing both secondary electrons and secondary positive ions. A fullerene beam current was measured with this FC and a high-aspect ratio FC, and measurement values were 3.3 nA and 4.5 nA, respectively. The difference of measurement values indicated that magnetic field was not enough to suppress secondary positive ions. Nevertheless, for install in beam transport line, it is notable that a fullerene beam current is able to measure without changing shape of a conventional FC. We will optimize magnetic field for a fullerene beam measurement in future.

Reference
4-53 Design of a Compact High-energy Focusing Lens System Combined with an Acceleration Tube

Y. Ishii a), T. Ohokubo a), J. Haga a), A. Kobayashi b) and S. Adachi b)

a) Department of Advanced Radiation Technology, TARRI, JAEA, b) Kobe Steel Ltd

A fine fabrication using high-energy focused proton beams, proton beam writing (PBW), is a promising technique for development of 3D structures such as optical devices and high sensitive sensors. High energy focused gaseous ion micro-/nanobeams are formed by the combination of a MV electrostatic accelerator, a beam line with a high resolution analyzing magnet and a high-energy focusing ion beam (MeV focused ion beam) system. Since each component combined in the system is longer than 10 m, the total size of its system reaches to several tens of meters. The wide usage of the MeV focused ion beam in various researches and development fields is, thereby, difficult due to the large size of the combination system.

The acceleration lens system, a series of acceleration lenses each of which has the both effects of focusing and accelerating ion beams in an electric field, was developed to achieve the demagnification over 1000 in JAEA. The focusing potential of the acceleration lens system was so far demonstrated by using the keV gaseous ion nanobeam system and forming the 170 nm H$_2^+$ beam at 50 keV$^1$.

On the other hand, a compact 1 MV focused ion beam system (compact 1 MV system) was developed using the combination of a 1 MV accelerator and a MeV focused ion beam system with magnetic quadruple lenses in Kobe Steel, LTD (KSL). Beam sizes with several micrometers in diameter are formed using the compact 1 MV system for the investigation of material element analysis in various materials using RBS techniques. JAEA in cooperation with KSL is developing a compact 2 MeV level gaseous ion nanobeam system aiming at 100 nm sized special resolution achieved by combining the acceleration lens system with the compact 1 MV system.

The MeV high demagnification lens system was designed using the acceleration lens system and an acceleration tube in the last year. The basic data of a multicusp ion source developed for another study were also measured to evaluate the conditions of beam current, energy and energy-spread required to its source.

The two kinds of performance of the acceleration lens and the acceleration tube needs to be investigated to design the compact MV high-energy focusing lens system. The performance data of the acceleration lens was a priori investigated from that of the keV gaseous ion nanobeam system.

The relation between focusing point and magnification was calculated employing Elkind’s equation applying accelerator tube parameters used in the compact 1 MV accelerator as a function of N value (accelerated beam energy/injection beam energy). The relation shows that the focusing points are formed in the accelerator tube when the N value of 27 and 20 for object points of 300 mm and 500 mm, respectively. Thereby, the usage of large demagnification, that is large N value, provide impossible to irradiate samples.

A 1.7 MeV focused proton with one hundred nm in diameter could be formed using the schematic design of the compact MV high-energy lens system as shown in Fig.1 with the parameters as listed in Table 1. The beam current at a focusing point is, however, small due to using a small aperture to inject small beam divergence angle into the accelerator tube. The one order smaller divergence beam angle injected into the accelerator tube is required to reduce aberrations as compared with the outgoing beam angle of the second acceleration lens. The acceleration lenses with a controllable outgoing beam divergence angle are investigating to increase the beam current at the focusing point. Furthermore, a multicusp ion source has been developing to increase beam current at the focusing point.

![Fig.1 A schematic design of a compact high-energy focusing lens system](image)

<table>
<thead>
<tr>
<th>Table.1 Parameters of the design</th>
</tr>
</thead>
<tbody>
<tr>
<td>voltage (kV)</td>
</tr>
<tr>
<td>------------</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>Incident angle (rad)</td>
</tr>
<tr>
<td>Outgoing angle (rad)</td>
</tr>
<tr>
<td>Magnification</td>
</tr>
<tr>
<td>Beam size</td>
</tr>
</tbody>
</table>

Reference
4-54 Micro-machining of Resists on Silicon by Proton Beam Writing: Part 2
(Fabrication of 3-D Structures Using a Negative resist)

N. Uchiya a), Y. Furuta a), H. Nishikawa a), J. Haga b), T. Satoh b), M. Oikawa b),
T. Ohkubo b), Y. Ishii b), T. Kamiya b) and S. Yamamoto c)

a)Department of Electrical Engineering, Shibaura Institute of Technology, b)Department of
Advanced Radiation Technology, TARRI, JAEA, c)Environment and Industrial Materials
Research Division, QuBS, JAEA

Proton beam writing (PBW) is a direct write technique for
the purpose of micro- or nano-meter sized machining 1). This
technique does not require an expensive mask for patterning
as well as electron beam though the mask is required for the
exposure with UV (EUV) and x-ray. The lateral straggling
of proton beam with energy of MeV order is far smaller
compared with electron beam. The depth of fabrication is
reached to several tens of micro-meter in a resist and
controlled by the beam energy. The PBW is a promising
technique for the micro-machining of thick resist and
three-dimensional (3-D) structures with high aspect ratios 2).

In our previous report 3), we confirmed the fabrication of
micro-structures with smooth and vertical side wall by the
PBW on PMMA and SU-8. In this paper we report the
results of fabrication of 3-D structures using two ranges of
proton beams in SU-8 layer controlled by the beam energies.
The micro-meter-sized structures by the PBW were
observed by a scanning electron microscope (SEM).

We used a layer of negative resist SU-8 (N-02 Wafer,
MEMS CORE) with a thickness of 50 µm on Si wafer.
The resist layer on silicon was exposed by focused proton
beam formed by the micro-beam system in TIARA, JAEA.
The beam size was estimated at about 1 µm in diameter
from secondary electron images of a commercially available
Ni mesh. The PBW was carried out with a resolution of
8192 x 8192 pixels in an area of 700 x 700 µm² by an
electrostatic scanner. The projected ranges of proton beam
into the resist layer were controlled by using two different
proton beam energies of 1.2- and 3.0-MeV. The fluence of
protons was controlled ranging from 20 to 80 nC/mm². 4)

In the first step, irradiation of the proton beam was
carried out at the energy of 3.0 MeV to form posts on the
substrate with a projected range of more than 50 µm. In the
next step, the irradiation was carried out at the lower energy
of 1.2 MeV to bridge across the posts at a depth of 28 µm.

After the development process for 3 minutes using a
developer for SU-8 and rinse in isopropyl alcohol, the
samples, which were coated with Pt by sputter to avoid
charging, were observed by the SEM.

The fabricated 3-D structures were shown in Fig.1. The
depth of the lines written by 1.2-MeV PBW were 22 µm
from the resist surface that was close to 21 µm estimated by
SRIM simulation 5). The vertical and smooth side wall of the
posts was formed by 3.0-MeV PBW. Using this technique,
we demonstrated fabrication of the structure like the Arch of
Triumph showed at Fig. 2.

The controlled ranges of the proton beam by changing the
beam energies enabled us to fabricate the multi-level
structures in the single resist layer. This shows the
capability of the PBW in the formation of 3-D
high-aspect-ratio structures.

References
3183 (1997) 128.
3) N. Uchiya et al., TIARA annual report 2005 (2006)
174.
405
5) J.F. Ziegler et al, The Stopping and Range of Ions in

Fig. 1 SEM image of the grid structure of SU-8
fabricated by PBW of energies of 1.2 and 3.0 MeV.

Fig. 2 SEM image of 3-D structure of SU-8 like
the Arch of Triumph fabricated by PBW at
energies of 1.2 and 3.0 MeV.
4-55 Measurement Technique of Two Dimensional Dose Distribution Using B3 Film Dosimeters and a PC-scanner

H. Hanaya, T. Agematsu and T. Kojima

Department of Advanced Radiation Technology, TARRI, JAEA

1. Introduction

Measurement of 2D dose distribution in large ion beam irradiation area is one of the indispensable techniques for R&D in material science and bio-technology using a cyclotron. Measurement technique for 2D distribution of relative ion beam fluence has been developed using Gaf-chromic films (HD-810) and an image scanner for a personal computer. This combined system has advantages such as costless, easiness of handling and speedy measurement with high spatial resolution. The measurable dose using this system is, however, not applicable to doses above 1200 Gy. The use of B3 film dosimeters (GEX B3002) was tested instead of Gaf-chromic films to expand measurable dose range of the combined system to higher dose region. The B3 dosimeters are commercially available in the aluminum package but would be used for measurement of ion beams after removing it, since ion beams have generally short penetration range. The optical readout of B3 dosimeters associates with heat treatment after irradiation for stabilization of dose response before readout. In the preliminary study, dose response of B3 films irradiated in the package by 2 MeV electron beams was linear up to 100kGy when readout using Green color component without heat treatment after irradiation. Dose responses of B3 film dosimeters irradiated with and without package were examined including their stability after irradiation.

2. Experimental and result

B3 film dosimeters with and without 200 μm-thick aluminum package were irradiated by 2 MeV electron beams to doses of 5 - 100 kGy. The irradiated dosimeters without heat treatment were readout using the image scanner (Canon LiDE50) with Green color components at different elapse times after irradiation.

The dose response curves for different elapse time after irradiation were shown in Fig.1: a) when dosimeters were kept in the package, and b) when dosimeters were not kept in the package. Any dose responses increase with dose and their curves are similarly linear up to at least 40 kGy. The dose responses increase with elapse time, but the slopes become gentle 24 h after irradiation, not depending on the use of the package.

3. Discussion

The B3 dosimeters show the linear dose response up to at least 40 kGy even used without package. It demonstrates that the dosimeters are usable also for ion beams having short penetration range. The dose response increases as a function of elapse time after irradiation. Such a variation can be minimized with heat treatment, but it becomes negligible order (< 3%) 24 h after irradiation without heat treatment.

The combined system with the B3 film dosimeters and an image scanner has a potential to be applied to measurement of 2D relative dose distribution of ion beams in the dose range of a few tens kGy, although further studies should be done using ion beams instead of electron beams.

References
Properties of Radiochromic Film Dosimeters for Low Energy Electron Beam

H. Seito a), T. Kojima a), T. Takei b), T. Ide b) and I. Mori b)

a)Department of Advanced Radiation Technology, TARRI, JAEA, b)IWASAKI ELECTRIC Co., Ltd

Introduction
Application of low energy electron accelerators (50-300 keV) is expanding in the field of radiation processing. Radiochromic film dosimeters have been used widely both in the industrial processes and research employing gamma-rays and electron beams1). These dosimeters have been calibrated commonly only for cobalt-60 gamma-rays, therefore the feasibility of this calibration should be studied also for their application to low energy electron beams.

Experimental
Two different radiochromic dosimeters were used in this study. One is nylon-based FWT602) with thickness of 10 μm, containing hexahydroxyethyl pararosaniline cyanide as the radiation-sensitive component. Another is GEX B3 radiochromic film dosimeter3). The B3 radiochromic dosimeter contains a radiochromic dye precursor of pararosaniline cyanide dissolved in polyvinyl butyral. Fundamental properties of these dosimeters were studied preliminarily for gamma-rays. The gamma-ray irradiation was carried out using a 60Co plaque source (2.6 PBq) at a dose rate of 10 kGy/h, which was measured with the ionization chamber periodically calibrated at AIST. Optical density was measured using Hitachi spectrophotometer model U-3310. Wavelength of 600 nm and 554 nm is used for dose measurement of FWT60 and B3, respectively.

Results and discussion
Response curves of the two different dosimeters are shown in Fig.1. The dose response of FWT60 is larger than that of B3. The response curve of FWT60 is nearly linear with dose up to 30 kGy and become sublinear at higher doses, on the other hand, that of B3 is nearly linear with dose up to 60 kGy.

The stability of absorbance is studied both for FWT60 and B3 after irradiation to doses of 15, 25, 35, 45, and 55 kGy, for 1 month. The variation of absorption occurred after irradiation is less than 2% for both dosimeters showing long shelf-life stability. These results should be a base for application of these radiochromic dosimeters to low energy electron beams.

References
2) A. Miller, Riso-M-2401. (1983)
**4-57 Standard Dosimetry of a few MeV Electron and $^{60}$Co Gamma-ray in Radiation Processing**

H. Seito, T. Kojima, H. Kaneko and N. Haneda

Department of Advanced Radiation Technology, TARRI, JAEA

**Introduction**

The high-dose calibration technique using a parallel-plate ionization chamber system has been developed in the JAEA dose calibration facility[1]. The facility equips two $^{60}$Co $\gamma$-ray plaque sources covering wide dose-rate range of 5-20000 Gy/h which have overlapping dose-rate region with that at the National Institute of Advanced Industrial Science and Technology (AIST), the national standard dosimetry laboratory in Japan. Consistency of dose calibration results at JAEA and those at AIST was verified without additional correction regarding to difference in characteristics of radiation fields involving current/exposure-rate conversion coefficients given in the dose rate level of 10-100 Gy/h at the AIST and high-dose rates of 5-20000 Gy/h at JAEA.

**Results and discussion on dose intercomparison**

The uncertainty associated with the established standard dose measurement procedure in JAEA has been reviewed. The total uncertainty in $^{60}$Co gamma-ray absorbed dose delivered by the JAEA calibration system is evaluated to be 2.2%(2 $\sigma$). The uncertainty of dose measurement using JAEA alanine/ESR transfer dosimeters is also estimated to be 3.4%(2 $\sigma$) for $^{60}$Co gamma-ray irradiation. A dose intercomparison study in the dose range of 1-50 kGy was carried out with National Physical Laboratory (NPL) to confirm our estimated uncertainty, employing transfer dosimeters: alanine dosimeters and dichromate dosimeters[2]. The results of dose intercomparison between JAEA and NPL show good agreement within about 2% in the dose range of 1-50 kGy. The results assure the uncertainties in gamma-ray absorbed dose measurement at JAEA using parallel-plate ionization chamber as a working reference dosimeter and alanine dosimeters as reference transfer dosimeters.

Dose comparison was performed for 2-MeV electrons employing cellulose triacetate (CTA) and alanine film dosimeters which were independently calibrated for 2-MeV electrons and $^{60}$Co gamma-rays using the calorimeter and the ionization chamber systems respectively[3]. Agreement of two different dose evaluations was better than 2.0%(1 $\sigma$) as shown in Fig 1. These results should enable us to achieve highly reliable dosimetry for the quality assurance in electron radiation processing on the basis of gamma-ray dose standards.

**Fig.1** Correspondence of electron dose evaluation (nominal dose) with gamma-ray dose evaluation (estimated dose)

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5-01 Safety Measures, Utilization Status and Spread of Research Results at TIARA Facility


Department of Advanced Radiation Technology, TARRI, JAEA

Periodical inspections per every month and three months were performed surely and safety of the facility was also maintained certainly. Several safety measures in the TIARA facility, such as the injection of sealing material to repair the cracked surface on the side wall of the building where leaking of rain sometimes occurred, the repair of cracked concrete surface on the higher ceiling and the exchange of ceiling fluorescent lamps in the second target room, the exchange of batteries in the emergency lamps, and so on, were carried out. Furthermore, a safety handrail on the roof and supplementary garden lights along a narrow walking trail as shown in Photo. 1 were also installed.

Photo. 1 Installation of safety handrail on the roof (left) and supplementary garden lights along narrow walking trail (right).

The shifts of utilization time of cyclotron and electrostatic accelerators assigned to the respective research fields are shown in Fig. 1 and Fig. 2. For recent 9 years, the total utilization time of cyclotron was constantly about 2500 h after it was about 2800 h for initial 5 years since 1993. However in fact, operation time of the cyclotron for above period was increased slightly to compensate time losses because the efficiency of the utilization gradually deteriorated by the frequent alteration of the operation condition. On the other hand, especially in 2006, utilization time of three electrostatic accelerators increased remarkably up to 491 days because of the increase of common use, although it was 440-460 days per year since 1995.

In regard to new utilization system introduced from last year, some issues were also improved. The application forms for research subjects and experimental planning was treated by an “electronic application system” which was newly constructed. The number of times for the application to research subjects per year was changed to once from twice. The designed percentage for internal, common and effective use in whole utilization frame was unified to 70:20:10 comprehensively for 4 accelerators.

The ratio of allotment time to application one for the first half period through 2007 was estimated to be 0.45 and 0.410 for cyclotron and electrostatic accelerators, respectively, which were the worst values so far because of the propose of reduction of appropriation. Actually, proposed budget was replenished rather enough after the allotment of utilization time. Therefore, utilization time for 2007 will be recovered to normal state.

The works for utilization and usher’s window were smoothly performed similarly to past years. The average number of persons for one month was 493 (38.4 for last three years) for staying, 52.5 (41.7) for the loan of glass budge and ID card, 41.9 (38.3) for the reception of facility utilization, 179 (115) for the short term visitors in TIARA facility, and so on.

The first Takasaki Advanced Radiation Research Symposium instead of the former TIARA Research Review Meeting was held successfully on June 22-23 at Takasaki city gallery. The number of presentations for oral and poster sessions was 21 and 158, respectively. Participants to this symposium were 412, which increased 85 persons compared to last meeting.

In addition, the JAEA Takasaki annual report 2005 was also issued firstly instead of TIARA annual report. This annual report summarized research results and activities which were done during fiscal year 2005 at Takasaki Advanced Radiation Research Institute. This report consists of 240 pages including several coloured sheets, and was printed as 700 books.

Electric power saving in TIARA facility during the “golden week” in Japan was achieved by the complete stop of large devices and equipments similar to a few past years.

Reference
5-02 Operation of AVF Cyclotron

T. Nara a), T. Agematsu a), I. Ishibori a), S. Kurashima a), K. Yoshida a), T. Yuyama a), T. Ishizaka a), S. Okumura a), N. Miyawaki a), H. Kashiwagi a), Y. Yuri a), W. Yokota a), K. Akaiwa b), T. Yoshida b), S. Ishiro b), Y. Arakawa b), T. Yoshida b), S. Kanou b), A. Ihara b) and K. Takano b)

a) Department of Advanced Radiation Technology, TARRI, JAEA. b) Beam Operation Service, Co., Ltd.

**Operation**

The AVF cyclotron was smoothly operated in fiscal 2006 and all the planned experiments were carried out without cancellation due to troubles relating to the cyclotron. The operation condition of the cyclotron is frequently changed because many kinds of ion species are required for the research in materials science and biotechnology with short irradiation time around an hour. Therefore the number of annual experiments reached 840. Table 1 shows the statistics for the cyclotron operation. The total operation time amounts to 3459.2 hours which is the longest so far.

<table>
<thead>
<tr>
<th>Statistics for cyclotron operation in fiscal 2006.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beam service time</td>
</tr>
<tr>
<td>Machine tuning</td>
</tr>
<tr>
<td>Beam development</td>
</tr>
<tr>
<td>Total operation time</td>
</tr>
<tr>
<td>Change of particle and/or energy</td>
</tr>
<tr>
<td>Change of beam course</td>
</tr>
<tr>
<td>Change of harmonic number</td>
</tr>
<tr>
<td>The number of experiments</td>
</tr>
<tr>
<td>Experiment canceled due to machine trouble</td>
</tr>
</tbody>
</table>

The percentages of operation time of the year used for experiments, joint-use, machine tuning, and beam development are 72.8%, 5.7%, 20.0%, and 1.5%, respectively.

A multi-cusp ion source was used to produce hydrogen ions. In the case of production of ions heavier than He, two ECR ion sources are used alternatively. Table 2 shows the ratios of operation time of each ion source. Figure 1 shows popularity of major ions used for experiments. The tendency of those statistics is similar to that of the past years.

**Table 2. Operation times of ion sources.**

<table>
<thead>
<tr>
<th>Ion source</th>
<th>Operation time</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Multi-cusp</td>
<td>1027.2 hr</td>
<td>26.7%</td>
</tr>
<tr>
<td>ECR (OCTOPUS)</td>
<td>1826.7 hr</td>
<td>47.5%</td>
</tr>
<tr>
<td>ECR (HYPERNANOGAN)</td>
<td>991.5 hr</td>
<td>25.8%</td>
</tr>
</tbody>
</table>

**Maintenance**

The number of the troubles of the year was 324, and they were quickly repaired without cancellation of experiments.

The regular yearly overhaul was carried out for five weeks in summer. About two hundreds of the rubber hoses which deteriorated in a high temperature and radiation environment for more than 15 years were renewed. The internal seals of about four hundreds Vebeo joints which were used for copper pipes of the trim-coils were replaced because cracks were found on a number of the seals.

The routine maintenance of power supplies for a week in autumn Most current power supplies were modified to maintain good performance for the next reason. A DAC amplifier of a current feedback control circuit in each power supply was enclosed in a constant-temperature box heated to 50°C by a thermostat-controlled oven for high current stability. However, breakdown of some parts in the DAC amplifier was increasing, and it was found that this temperature hastened deterioration of the parts. Therefore, the temperature has been changed to 25°C which is kept by a Peltier device. The stability of intensity and position of beam has been improved as a result, which helps to shorten the time of beam tuning for the microbeam formation.

**Technical development**

The beam acceleration tests were successfully carried out for 14 MeV H+, 39 MeV 28Si10+ and 60MeV 11B3+, and they are ready to be used for experiments.

The development of a multipole magnets system for uniform-irradiation has been started. It allows exactly simultaneous beam irradiation over a wide area at high uniformity1), which is impossible by means of raster scanning of a spot beam.

The main beam-probe of the cyclotron has been modified to measure a beam phase width using a plastic scintillator and a test experiment was successfully done.

**Reference**

5-03 Operation of the Electrostatic Accelerators

K. Mizuhashi a), S. Uno a), K. Ohkoshi a), A. Chiba a), K. Yamada a), Y. Saitoh a), Y. Ishii b), T. Satoh a), W. Yokota a), T. Kitano b), T. Takayama b), T. Orimo b), S. Kanai b), A. Ohmae b), M. Kouka b) and Y. Aoki b)

a) Department of Advanced Radiation Technology, TARRI, JAEA, b) Beam Operation Service, Co., Ltd

1. Status of electrostatic accelerators

Three electrostatic accelerators of the TIARA facility operated smoothly in fiscal 2006 and all the planned experiments were carried out, except those canceled by users. The operating times of the tandem accelerator, the single-ended accelerator and the ion implanter were 1959.6, 2498.6 and 1898.9 hours respectively. These are almost same as the past years but the number of operating day increased. The total operating times since operation started are 24821, 29585 and 23226 hours, respectively. Figure 1 shows the total time of each accelerator from 1991 to 2006.

The number of the troubles was small compared with the past years. The troubles of the year are characterized by the failure of condensers. Many condensers are used in the power supplies and the computers of the accelerator system. A lifetime of a condenser is generally about 10 years, but most of power supplies of the TIARA facility are used over 15 years. Main troubles of condenser are as follows. The condensers used in the computers of the tandem accelerator control system for seven years failed and a number of the abnormal condensers were found in three of six control computers. The condensers used in the computers of the tandem accelerator control system for seven years failed and a number of the abnormal condensers were found in three of six control computers. The single-end accelerator was stopped for two days since the condenser of the RC oscillation circuit for the RF ion source was shorted. All the lost beam time due to the troubles was compensated in the year.

2. Improvement for the accelerators

The sliding mechanism for one of a couple of the chain motors of the tandem accelerator, which give constant tension to the pellet chain, was modified to move more lightly. This drastically reduced the dust production by the chain rotation at high speed 1), and the ripple of the high voltage terminal attributed to the chain vibration lowered by about 20% due to better stability of the chain rotation. As to single-ended accelerator, the damage of the plastic electric insulation support for the RF-electrode frequently occurred because of the heating by the electric discharge occurred at the bubble parts in the support. In order to prevent the damage, the supports made of polypropylene were replaced with those of polycarbonate after checking that they had no bubbles by eyes through their transparency.

The cluster ions of C and Au are accelerated by the tandem accelerator, but most (more than 90%) of clusters are broken up by the collision with the charge exchange gas N₂. Therefore, the gas stripper system was modified so as to measure the charge exchange efficiency for various gas species. The results of the measurement indicate that a gas with a larger molecular weight gives the higher charge exchange efficiency 2).

3. New beam development

As to the ion implanter, the Ge ion, which was requested by the users of the inorganic functional material research, was newly accelerated. It is difficult to stably generate ions of a low melting-point material, such as Ge, with a Freeman ion source because it easily runs from the material holder. Therefore, the holder has been modified so that the short of the arc circuit by the melted material and its loss should not take place. Stable Ge ion beam was successfully generated as a result and about 5μA was accelerated.

References
1 Individual monitoring

(1) Individual monitoring for the radiation workers

Table 1 shows a distribution of effective dose of the radiation workers in FY 2006. The effective dose values of almost all radiation workers were below the detection limit (0.1 mSv).

The maximum dose was 0.6 mSv/y due to the overhaul of the TIARA AVF cyclotron.

Table 1. Distributions of the effective dose of the radiation workers in FY 2006.

<table>
<thead>
<tr>
<th>Distribution range of effective dose (mSv)</th>
<th>Number of persons in each period (Person)</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of persons under radiation control (A)</td>
<td>512 561 546 567 631</td>
<td></td>
</tr>
<tr>
<td>0.1 ≤ HE ≤ 1.0</td>
<td>0 8 2 0 9</td>
<td></td>
</tr>
<tr>
<td>1.0 ≤ HE ≤ 5.0</td>
<td>0 0 0 0 0</td>
<td></td>
</tr>
<tr>
<td>5.0 ≤ HE ≤ 15.0</td>
<td>0 0 0 0 0</td>
<td></td>
</tr>
<tr>
<td>15.0 &lt; HE ≤ 60.0</td>
<td>0 0 0 0 0</td>
<td></td>
</tr>
</tbody>
</table>

Table 2. Number of persons who temporarily entered the radiation controlled areas in FY 2006.

<table>
<thead>
<tr>
<th>Periods</th>
<th>1st quarter</th>
<th>2nd quarter</th>
<th>3rd quarter</th>
<th>4th quarter</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of persons</td>
<td>423</td>
<td>545</td>
<td>622</td>
<td>491</td>
<td>2083</td>
</tr>
</tbody>
</table>

(2) Individual monitoring for the visitors and others

Table 2 shows number of persons who temporally entered the radiation controlled areas. The effective dose of all persons was less than 0.1 mSv.

Table 3. Monitoring results of released radioactive gases and dust in FY 2006.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Maximum concentration (Bq/cm³)</th>
<th>Activity (Bq)</th>
<th>Number of persons under radiation control (A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>41Ar</td>
<td>1.4×10⁻⁶ 1.4×10⁻⁶ 1.5×10⁻⁶ 1.8×10⁻⁶ 1.8×10⁻⁶</td>
<td>1.2×10⁷ 3.8×10⁷ 1.7×10⁷ 8.6×10⁷ 1.5×10⁷</td>
<td>512 561 546 567 631</td>
</tr>
<tr>
<td>11C</td>
<td>1.4×10⁻⁶ 1.4×10⁻⁶ 1.5×10⁻⁶ 1.5×10⁻⁶ 1.5×10⁻⁶</td>
<td>5.9×10⁷ 1.8×10⁷ 1.5×10⁷ 1.2×10⁷ 1.1×10⁷</td>
<td>512 561 546 567 631</td>
</tr>
<tr>
<td>13N</td>
<td>Maximum concentration (Bq/cm³)</td>
<td>Activity (Bq)</td>
<td>Number of persons under radiation control (A)</td>
</tr>
<tr>
<td>18F</td>
<td>Maximum concentration (Bq/cm³)</td>
<td>Activity (Bq)</td>
<td>Number of persons under radiation control (A)</td>
</tr>
<tr>
<td>18F</td>
<td>Maximum concentration (Bq/cm³)</td>
<td>Activity (Bq)</td>
<td>Number of persons under radiation control (A)</td>
</tr>
<tr>
<td>65Zn</td>
<td>Maximum concentration (Bq/cm³)</td>
<td>Activity (Bq)</td>
<td>Number of persons under radiation control (A)</td>
</tr>
</tbody>
</table>

3 Monitoring for external radiation and surface contamination

External radiation monitoring was routinely carried out in around the radiation controlled areas and surface contamination monitoring was also carried out. Neither unusual value of dose equivalent rate nor surface contamination were detected.

Figure 1 shows a typical example of distribution of the dose equivalent rate at the radiation controlled area in the cyclotron building.

Fig. 1. Dose equivalent rate distribution at the radiation controlled area in the cyclotron building.

Measurement date: 27th March 2007
Measuring position: Indicated with × 1m above floor
Unit: μSv/h (the dose equivalent rate less than 0.2 μSv/h is not indicated)
1. The radioactive wastes management

The radioactive wastes generated in TIARA are managed by Utilities and Maintenance Section. The main radioactive wastes are the solid wastes generated by the research experiment and the maintenance of the cyclotron. Other radioactive wastes are the liquid wastes such as inorganic waste fluids generated from the research experiment and the air-conditioning machines in controlled area. These wastes are managed according to properties.

2. Solid radioactive waste

Table 1 shows the amounts of solid wastes at various properties and kinds generated in each quarter of FY 2006. The main solid waste is generated by the research experiment and the maintenance of the cyclotron.

Combustible wastes are rubber gloves, paper, and clothes, etc. Incombustible wastes are metal pieces, the glasses, and contaminated parts.

3. Liquid radioactive waste

Table 2 shows the amounts of liquid wastes generated in each quarter of FY 2006. Liquid waste was almost inorganic waste water generated with chemical experiments and operation of air conditioning units installed in each room of the first class radiation controlled area. Larger quantity of waste water in summer season (2nd quarter) is mainly due to condensed water, which is treated by evaporation, and inorganic water is reused in the controlled area. Only small amounts of concentrated liquid are generated by the evaporation.

Table 1. Radioactive solid wastes generated in FY 2006.

<table>
<thead>
<tr>
<th>Items</th>
<th>Amounts of generation in each periods (m³)</th>
<th>Number of package /drum</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1st quarter</td>
<td>2nd quarter</td>
</tr>
<tr>
<td>Category A*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1)Combustible</td>
<td>0.38</td>
<td>0.44</td>
</tr>
<tr>
<td>2)Incombustible</td>
<td>0.08</td>
<td>0</td>
</tr>
<tr>
<td>Compressible</td>
<td>0.08</td>
<td>0</td>
</tr>
<tr>
<td>Filters</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Incompressible</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Ion exchange resin</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Category B*</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

* defined by dose at the outer surface of container: (A) < 2 mSv/h ≤ (B)
** 200-liter drum

Table 2. Radioactive liquid waste generated in FY 2006.

<table>
<thead>
<tr>
<th>Items</th>
<th>Amounts of generation in each periods (m³)</th>
<th>Number of package /drum</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1st quarter</td>
<td>2nd quarter</td>
</tr>
<tr>
<td>Category A*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1)Inorganic</td>
<td>10.42</td>
<td>16.90</td>
</tr>
<tr>
<td>2)Organic</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Organic</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Oil</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>3)Sludge</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>4)Evaporation residue</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Category B*</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

* defined by concentrations in Bq/cm³(β,γ): (A) < 3.7×10⁻³ ≤ (B) < 3.7×10⁴
5-06 Utilization of the Electron Accelerator and Gamma-ray Irradiation Facilities

T. Kojima a), H. Kaneko a), N. Haneda a), H. Hanaya a), R. Yamagata a), H. Seito a), T. Kanazawa a), S. Koyama b), T. Yamaguchi b), I. Kawashima b), N. Yagi b) and M. Takagi b)

a) Department of Advanced Radiation Technology, TARRI, JAEA , b) Radiation Application Development Association, RADA

1. Present status of the utilization of electron accelerator and gamma-ray irradiation facilities

An electron accelerator and three gamma ray irradiation facilities were operated for various research subjects as operation plan in FY 2006 without serious trouble. Distribution of research fields and number of experiment subjects are shown in Fig.1 and Fig.2, respectively. The number of research subjects for the electron accelerator, the cobalt first facility, the cobalt second facility, and the food irradiation facility, is 544, 159, 622, and 357, respectively. Those for gamma-ray irradiation facilities in FY 2006 are lower than those in FY 2005, while it shows the increase of longer-time irradiation for radiation-resistance tests of materials and apparatus used in nuclear power plants or space.

Fig.1 Distribution of research fields (FY 2006).

Fig.2 Number of research subjects (FY 2002-2006).
5-07 Operation of the Electron Accelerators and 
Gamma-ray Irradiation Facilities

H. Kaneko a), H. Hanaya a), N. Haneda a), R. Yamagata a), H. Seito a), T. Kanazawa a),
T. Kojima a), S. Koyama b), T. Yamaguchi b), I. Kawashima b), N. Yagi b) and M. Takagi b)

a)Department of Advanced Radiation Technology, TARRI, JAEA, b)Radiation 
Application Development Association, RADA

1. Operation

1.1 Electron accelerator

The electron accelerator was on service without serious 
trouble in 9:00~17:30 on Monday and Friday, and in 
8:30~23:00 on Tuesday to Thursday, to satisfy demand of 
operation time from users.

The annual operation time for the electron accelerator, 
as shown in Fig.1, is 597.4 h (517 h for vertical beams, 
80.4 h for horizontal beams). That for the accelerator 
No.2 operated until 2004 is also shown in Fig.1. Total 
operation time of the accelerator No.1 in FY2006 is about 
30% lower than that in FY2005, but the number of 
research subject in FY2006 is the same as that in FY2005 
indicating the increase of short time irradiation 
experiments. The accelerator served mainly for 
graft-polymerization for new material development, 
radiation effect study on semiconductors, and various 
experiments of visiting users.

1.2 Gamma-ray irradiation facilities

The Co-60 gamma-ray irradiation facilities consist of 
three buildings (8 rooms) and cover a wide dose-rate 
range from 0.04 Gy/h to 20 kGy/h with eight irradiation 
cells. The annual operation time for the cobalt first, 
second irradiation facilities and food irradiation facility is 
20,844 h, 6,335 h and 3,150 h, respectively, as shown in 
Fig.2.

The first irradiation facility served mainly for 
radiation-resistance testing of cables used in nuclear 
power plants and various materials used in J-PARC 
facility with long irradiation period. The second 
irradiation facility served mainly

![Fig.1 Sift of annual operation time of electron accelerators](image1)

![Fig.2 Sift of annual operation of Co gamma-ray irradiation facilities](image2)

for development of new functional materials and other 
research subjects of visiting users, involving irradiation 
room No.6 operated as hourly scheduled. The food 
irradiation facility served mainly for development of 
detection method for irradiated foods and radiation 
resistance testing at lower dose rates.

2. Maintenance

2.1 Electron accelerator

The operation of the accelerator was interrupted for 10 
days in October, 2006 and 5 days in March 2007 because of 
periodical maintenance check of accelerator body and 
icidental equipment, e.g. conveyor and doors of 
irradiation room and repair of current measurement 
system at the horizontal beam line.

2.2 Gamma-ray irradiation facilities

The periodical maintenance check mainly of mechanical 
system for radiation source transportation has been 
performed every three year among three gamma-ray 
irradiation facilities one by one. The periodical check mainly 
of interlock system has been performed two times a year for 
all the facilities.

The maintenance check of the cobalt first irradiation 
facility was done in July 2006, with 18 days interruption. 
The control system for the cobalt second irradiation facility 
was 18 years old and therefore waiting for replacement of 
the sequencer for operation.

Co-60 sources were purchased and loaded to the 
irradiation room No.6 in the second irradiation facility to 
maintain total activities of the room. The old waste 
sources of about 30 pieces were disposed through JRIA.
COMMON USE PROGRAM in Takasaki

M. Otsubo, M. Hoto and R. Suzuki

Department of Advanced Radiation Technology, TARRI, JAEA

1. Introduction

The facilities of JAEA is widely opened to users like universities, public institutes, and the industries, etc. COMMON USE PROGRAM, the system of facility use for the user’s purpose on fee-charging basis, started since 2006 taking over the former system. There are 3 OPEN USE facilities in Takasaki; Co-60 Gamma-ray Irradiation Facilities, Electron Accelerator and TIARA. In addition to these, some of the off-line devices can be used.

We round up the Research Proposals dividing one year into two terms, and the execution possibility and the validity of the experimental plan are checked by the special committee. The facility usage fee will be the total of the admission fee, irradiation fee and additional charge. In case of Non-proprietary research, the users must report the irradiation result to JAEA, and could be exempt from irradiation fee. JAEA will open the reports to the public. It is also possible for universities to apply from the facility use program of Tokyo University. Those applications are accepted as priority case. Table 1 shows the outline of the program.

Table 1 Classification of COMMON USE PROGRAM

<table>
<thead>
<tr>
<th>Purpose</th>
<th>research and development</th>
<th>except R&amp;D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Classification</td>
<td>general</td>
<td>priority case</td>
</tr>
<tr>
<td>Result</td>
<td>non-proprietary</td>
<td>proprietary</td>
</tr>
<tr>
<td>Application</td>
<td>regular&amp; irregular</td>
<td>regular</td>
</tr>
<tr>
<td>Referee</td>
<td>yes</td>
<td>no</td>
</tr>
<tr>
<td>Charge*</td>
<td>A</td>
<td>B</td>
</tr>
</tbody>
</table>

*A= admission fee + additional charge
B= admission fee + irradiation fee + additional charge
C= admission fee + irradiation fee + depreciation

2. Public relations

The information like an outline of this system, guidelines for applicants, format download, etc can be seen on JAEA website(http://www3.tokai-sc.jaea.go.jp/sangaku/3-facility/index.html). In addition to this, we go out for publicity and to search current needs of the outside of JAEA. In 2006, we participated in 15th Polymer Material Forum(Osaka), Seminar for advancement of RI/radiation use(Nagoya), Opening Symposium of Center for Flexible Micro machining(Tokyo), and so on.

3. Use in 2006

There were 28 Research Proposals in 2006 at Takasaki, and 16 of all hoped to use with open result. Including the users from priority case, we accepted 325 applications from 54 users and half of the users were new. We can find that the percentage of users from universities exceeds the others except Co-60 Facility from Table 2. However we expect there should be latent needs in the industries. Therefore we are trying to find them by the public relations as above-mentioned and also going to start another new system.

Figure 1 shows classification of all 325 applications. On TIARA, Non-proprietary use represents more than 50%, while Co-60 Facility and Electron Accelerator have a few%. This tendency may come from the user’s affiliation.

Table 2 User’s affiliation of each facility

<table>
<thead>
<tr>
<th>Facility</th>
<th>User</th>
<th>Public Institute</th>
<th>Others</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>AVF Cyclotron</td>
<td>5</td>
<td>2</td>
<td>3</td>
<td>10</td>
</tr>
<tr>
<td>3MV Tandem Accelerator</td>
<td>3</td>
<td>0</td>
<td>2</td>
<td>5</td>
</tr>
<tr>
<td>3MV Single-ended Accelerator</td>
<td>1</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>400kV Ion Implanter</td>
<td>6</td>
<td>0</td>
<td>1</td>
<td>7</td>
</tr>
<tr>
<td>Co-60 Gamma-ray Irradiation Facility</td>
<td>5</td>
<td>0</td>
<td>12</td>
<td>17</td>
</tr>
<tr>
<td>Electron Accelerator</td>
<td>8</td>
<td>0</td>
<td>6</td>
<td>14</td>
</tr>
<tr>
<td>Total</td>
<td>28</td>
<td>2</td>
<td>24</td>
<td>54</td>
</tr>
</tbody>
</table>

Fig.1 Percentage of each classification

* the number of application
# Appendix

<table>
<thead>
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<th>Appendix</th>
<th>Page</th>
</tr>
</thead>
<tbody>
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<td>A1.2 Publication in Proceedings</td>
<td>229</td>
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<td>237</td>
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<td>240</td>
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<tr>
<td>Appendix 4 Type of Research Collaboration</td>
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<tr>
<td>Symbol used in the Appendix</td>
<td>243</td>
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</tbody>
</table>
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Appendix 1. List of Publication

A1.1 Publications in Journal

06J001 1-06 I
F. Takano, W. Wang, H. Akinaga, H. Ofuchi, S. Hishiki and T. Ohshima,
“Characterization of Mn-doped 3C-SiC prepared by ion implantation”

06J002 1-06 E
“(Nitrogen-Vacancy)-Complex Formation in SiC: Experiment and Theory”

06J003 1-06 I, G
K. K. Lee, M. Laube, T. Ohshima, H. Itoh and G. Pensl,
“Hall Effect and Admittance Measurements of n-channel 6H-SiC MOSFETs”

06J004 1-06 T, I
“Degradation of Charge Collection Efficiency Obtained for 6H-SiC n+p Diodes Irradiated with Gold Ions”

06J005 1-06 T, I
T. Ohshima, T. Satoh, M. Oikawa, S. Onoda, T. Hirao and H. Itoh,
“Charge Induced in 6H-SiC PN Diodes by Irradiation of Oxygen Ion Microbeams”

06J006 1-06 E
“Deactivation of nitrogen donors in silicon carbide”

06J007 1-06 I, S
“Structural Analysis of (Ga,Mn)N Epilayers and Self-Organized Dots using MeV Ion Channeling”

06J008 1-07 E
T. Umeda, J. Isoya, T. Ohshima, N. Morishita, H. Itoh and A. Gali,
“Identification of positively charged carbon antisite-vacancy pairs in 4H-SiC”

06J009 1-07 E
T. Umeda, N. Morishita, T. Ohshima, H. Itoh and J. Isoya,
“Electron paramagnetic resonance study of carbon antisite-vacancy pair in p-type 4H-SiC”
06J010  1-07  T
J. Isoya, M. Katagiri, T. Umeda, N. T. Son, A. Henry, E. Janzén, T. Ohshima and H. Itoh,
“Shallow phosphorus donors in 3C-, 4H-, and 6H-SiC”

06J011  1-07  E TGE
T. Umeda, N. T. Son, J. Isoya, N. Morishita, T. Ohshima, H. Itoh and E. Janzén,
“Electron paramagnetic resonance of the HEI4/SI5 center in 4H-SiC”

06J012  1-07  E
N. T. Son, T. Umeda, J. Isoya, A. Gali, M. Bockstedte, B. Magunsson, A. Ellison, N. Morishita, T. Ohshima, H. Itoh and E. Janzén,
“Divacancy model for P6/P7 centers in 4H- and 6H-SiC”

06J013  1-08  E
H. Matsuura, S. Kagamihara, Y. Itoh, T. Ohshima and H. Itoh,
“Mechanisms of reduction in hole concentration in Al-doped 4H-SiC by electron irradiation”

06J014  1-08  E
H. Matsuura, S. Kagamihara, Y. Itoh, T. Ohshima and H. Itoh,
“Relationship between defects induced by irradiation and reduction of hole concentration in Al-doped 4H-SiC”

06J015  1-09  O
Miyashita, T. Ohnuma, M. Iwasawa, H. Tsuchida and M. Yoshikawa,
“Generation of amorphous SiO2/SiC interface structure by the first-principles molecular dynamics simulation”

06J016  1-09  O
T. Ohnuma, A. Miyashita, M. Iwasawa, M. Yoshikawa and H. Tsuchida,
“Dynamical simulation of SiO2/4H-SiC(0001) interface oxidation process: from first-principles”

06J017  1-10  I
“Fluorescence EXAFS study on local structures around Eu atoms implanted in AlxGa1-xN”

06J018  1-10  I
A. Wakahara
“IMPACT OF AlGaN ON LUMINESCENCE CAPABILITY OF RARE?EARTH IONS IN AlGaN”

06J019  1-27  C, E, G
T. Yamaki, Y. Kozone, A. Hiroki, M. Asano, H. Kubota and M. Yoshida,
“Nano-structure controlled polymer electrolyte membranes for fuel cell applications prepared by ion beam irradiation”
ESC Transaction, 3 (2006) 103-112.
06J020 1-27 C, E, G
T. Yamaki, Y. Kozone, A. Hiroki, K. Hosoi, M. Asano, H. Kubota and M. Yoshida,
“Proton exchange membranes for fuel cell applications prepared by ion track technology”

06J021 1-28 E, G
J. Chen, M. Asano, Y. Maekawa and M. Yoshida,
“Suitability of some fluoropolymers used as base films for preparation of polymer electrolyte fuel cell membranes”

06J022 1-28 E, G
J. Chen, M. Asano, T. Yamaki and M. Yoshida,
“Preparation and characterization of chemically stable polymer electrolyte membranes by radiation-induced copolymerization of four monomers into ETFE films”

06J023 1-28 E, G
J. Chen, M. Asano, T. Yamaki and M. Yoshida,
“Chemical and radiation crosslinked polymer electrolyte membranes prepared from radiation-grafted ETFE films for DMFC applications”

06J024 1-28 E, G
J. Chen, M. Asano, Y. Maekawa, T. Sakamura, H. Kubota and M. Yoshida,
“Preparation of ETFE-based fuel cell membranes by UV-induced photografting and electron beam-induced crosslinking techniques”

06J025 2-03 E
N. Ito, A. Mase, N. Seko, M. Tamada, E. Sakata and Y. Kogi,
“Surface Treatment of Poly(tetrafluoroethylene) and Perfluoroethylene-propylene by Radiation Grafting”

06J026 2-04 E
A. Iwanade, T. Nomoto, D. Umeno, K. Saito and T. Sugo,
“Protein Binding Characteristics of Amphoteric Polymer Brushes Grafted onto Porous Hollow-Fiber Membrane”
J. Ion Exch., accepted.

06J027 2-06 E
Y. Wada, M. Tamada N. Seko and H. Mitomo,
“Emulsion grafting of vinyl acetate onto pre-irradiated poly(3-hydroxybutyrate) film”

06J028 2-07 E, G
N. Seko, Y. Ueki, H. Hoshina and M. Tamada,
“Preparation of graft adsorbent having amine groups and its Au(III) adsorption performance”
J. Ion Exch., in press.

06J029 2-08 E
H. Hoshina, N. Seko, Y. Ueki and M. Tamada,
“Synthesis of graft adsorbent with N-methyl-D-glucamine for boron adsorption”
J. Ion. Exch., in press.

06J030 2-09 E
Md. R. Awual, A. Jyo, M. Tamada and A. Katakai,
“Zirconium(IV) loaded bifunctional fiber containing both phosphonate and sulfonate as
arsenate adsorbent“

06J031 2-13 E
Y. Ueki, N. Seko, H. Hoshina and M. Tamada,
“Preparation of Polylactic Acid Nonwoven Fabric-based Metal Adsorbent by Radiation-Induced Graft Polymerization”
J. Ion. Exch., in press.

06J032 2-17 E, G
T. M. Quynh, H. Mitomo, N. Nagasawa, Y. Wada, F. Yoshii and M. Tamada,
“Properties of crosslinked polylactides (PLLA & PDLA) by radiation and its biodegradability”

06J033 2-18 E, G, O
"Preparation and characterization of CMC gels"

06J034 2-23 E,G
A. Kimura, M. Taguchi, Y. Ohtani, Y. Shimada, H. Hiratsuka and T. Kojima,
“Treatment of wastewater having estrogen activity by ionizing radiation”

06J035 3-03 C, G
Y. Yokota, S. Yamada, Y. Hase, N. Shikazono, I. Narumi, A. Tanaka and M. Inoue,
“Initial yields of DNA double-strand breaks and DNA fragmentation patterns depend on linear energy transfer in tobacco BY-2 protoplasts irradiated with helium, carbon and neon ions”

06J036 3-06 C
H. Yamaguchi, T. Morishita, K. Degi, A. Tanaka, N. Shikazono and Y. Hase,
“Effect of Carbon-ion Beams Irradiation on Mutation Induction in Rice”

06J037 3-20 C
Y. Matuo, S. Nishijima, Y. Hase, A. Sakamoto, A. Tanaka and K. Shimizu,
“Specificity of mutations induced by carbon ions in budding yeast Saccharomyces cerevisiae”

06J038 3-23 C
“Alteration of black Sigatoka (Mycosphaerella fijiensis Morelet) resistance in banana by in vitro irradiation using carbon ion-beams”

06J039 3-26 G
K. Satoh, H. Ohba, H. Sghaier and I. Narumi,
“Down-regulation of radioreistance by LexA2 in Deinococcus radiodurans”

06J040 3-30 T
K. Akamatsu,
“A novel methodology for characterizing strand-break termini and damaged bases in plasmid DNA exposed to ionizing radiation”

06J041 3-31 C
A. Urushibara, N. Shikazono, R. Watanabe,
K. Fujii, P. O'Neill and A. Yokoya,
“DNA damage induced by the direct effect of He ion particles”

06J042 3-33 C
H. Matsumoto, A. Takahashi and T. Ohnishi,
“Nitric oxide radicals choreograph a radioadaptive response”

06J043 3-33 C
H. Matsumoto, H. Hamada, A. Takahashi,
Y. Kobayashi and T. Ohnishi,
“Vanguards of paradigm shift in radiation biology: Radiation-induced adaptive and bystander responses”

06J044 3-33 C
N. Hamada, H. Matsumoto, T. Hara and Y. Kobayashi,
“Intercellular and intracellular signaling pathways mediating ionizing radiation-induced bystander effects”

06J045 3-33 C
Z.-G. Cui, T. Kondo and H. Matsumoto,
“Enhancement of apoptosis by nitric oxide released from α-phenyl-tert-butyl nitrone under hyperthermic conditions”

06J046 3-34 C
N. Hamada, T. Hara, T. Funayama, T. Sakashita and Y. Kobayashi,
“Energetic heavy ions accelerate differentiation in the descendants of irradiated normal human diploid fibroblasts”

06J047 3-34 C
N. Hamada, H. Matsumoto, T. Hara and T. Y. Kobayashi,
“Intercellular and intracellular signaling pathways mediating ionizing radiation-induced bystander effects”

06J048 3-34 C
N. Hamada, T. Funayama, S. Wada, T. Sakashita,
T. Kakizaki, M. Ni and Y. Kobayashi,
“LET-dependent survival of irradiated normal human fibroblasts and their descendents”

06J049 3-34 C
N. Hamada, G. Schettino, G. Kashino, M. Vaid,
K. Suzuki, S. Kodama, B. Vojnovic, M. Folkard,
M. Watanabe, B.D. Michael and K.M. Prise,
“Histone H2AX phosphorylation in normal human cells irradiated with focused ultrasoft X-rays: Evidence for chromatin movement during repair”

06J050 3-39 C, T
M. Hino, S. Wada, Y. Tajika, Y. Morimura,
N. Hamada, T. Funayama, T. Sakashita,
T. Kakizaki, Y. Kobayashi and H. Yorifuji,
“Heavy ion microbeam irradiation induces ultrastructural changes in isolated single fibers of skeletal muscle”
K. Akeo, T. Funayama, A. Ogawa, N. Hamada, Y. Akeo and Y. Kobayashi,
“Effects of gamma irradiation on bel-2 and p53 expression in the porcine ciliary body”

“Distinct modes of cell death by ionizing radiation observed in two lines of feline T-lymphocytes”

“Killing of feline T-lymphocytes by gamma-rays and energetic carbon ions”

“Vulnerability of feline T-lymphocytes to charged particles”

M. Watanabe, T. Sakashita, A. Fujita, T. Kikawada, D.D. Horikawa, Y. Nakahara, S. Wada, T. Funayama, N. Hamada, Y. Kobayashi and T. Okuda,
“Physiological changes leading to anhydrobiosis improve radiation tolerance in Polypedilum vanderplanki larvae”

E. Ling, K. Shirai, R. Kanekatsu, K. Kiguchi, Y. Kobayashi, T. Funayama and H. Watanabe
“Contribution of circulating hemocytes to the regeneration of heavy ion beams (^{12}\text{C}^{5+}) irradiated hematopoietic organs in the silkworm, Bombyx

M. Watanabe, T. Sakashita, A. Fujita, T. Kikawada,
D.D. Horikawa, Y. Nakahara, S. Wada,
T. Funayama, N. Hamada, Y. Kobayashi and T. Okuda,
“Biological effects of anhydrobiosis in an African chironomid, Polypedilum vanderplanki on radiation tolerance”
mori, through the way of phagocytosis of injured cells after invasion”

06J060 3-48 C
“Development of the irradiation method for the first instar silkworm larvae using locally targeted heavy-ion microbeam”

06J061 3-52 G
M. Ukai and Y. Shimoyama,
“Free radicals in irradiated wheat flour detected by electron spin resonance”

06J062 3-53 O, G
T. Fujiwara, T. Yamaoka, Y. Kimura and K. J. Wynne,
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“DEVELOPMENT OF THE CENTRAL REGION FOR CONTROL OF A BEAM PHASE WIDTH AT THE JAEA AVF CYCLOTRON”

06C059 4-54 S, T
N. Uchiya, T. Harada, M. Murai, H. Nishikawa, J. Haga, T. Satoh, Y. Ishii and T. Kamiya,
“Micro-machining of resists on silicon by proton beam writing”

06C060 4-54 S, T
打矢直之、古田祐介、西川宏之、芳賀潤二、佐藤隆博、石井保行、神谷富裕
“集束プロトンビーム描画による厚膜レジストの微細加工”
秋季第 67 回応用物理学会、p.649、2006 年 8 月(滋賀)

06C061 4-54 S, T
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06C062 4-54 S, T
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“集束プロトンビームによる高アスペクト比微細加工”
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06C063 4-54  S, T
古田祐介、打矢直之、西川宏之、芳賀潤二、
及川将一、佐藤隆博、石井保行、神谷富裕、
“集束プロトンビーム描画による三次元構造体
の作製”
春季第 54 回応用物理学会、p.780、2007 年 3
月(神奈川).

06C064 4-57  E
T. Kojima, H. Sunaga and R. Tanaka,
“Consistency in evaluation of a few MeV electron
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Appendix 2

List of Related Patents

06PAT001 1-20
井岡郁夫（原子力機構）他 神戸製鋼所、コベルコ研との共同出願、
「耐粒界腐食性および耐応力腐食割れに優れたオーステナイト系ステンレス鋼およびその製造方法」
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06PAT002 1-27
高木、小林、斎藤（トヨタ自動車）、吉田、八巻、浅野（原子力機構）
「Functional membrane and electrolyte membrane for fuel cell and method for producing the same」，PCT/JP2006/303504

06PAT003 1-27
織木、八巻、浅野、吉田（原子力機構）
「プロトン伝導度測定方法と装置」
特願 2006-043562 号

06PAT004 1-27
小林（トヨタ自動車）、八巻、浅野、吉田、前川（原子力機構）
「機能性膜の製造方法、及び燃料電池用電解質膜の製造方法」
特願 2006-317281 号

06PAT005 2-3
瀬古、玉田（原子力機構）、伊藤、坂田（九州日立マクセル）
「高周波基盤およびその製造方法」
特願 2005-359923 号

06PAT006 2-7
瀬古、玉田、吉井（原子力機構）
「金を吸着回収する吸着材の合成方法及び排水処理」
特開 2005-154973 号

06PAT007 2-16
玉田、八木、長澤（原子力機構）金澤（住友電工ファインポリマー）
「ポリ乳酸製熱収縮材」
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06PAT008 2-16
金澤、川野（住友電工ファインポリマー）、玉田、長澤、八木（原子力機構）
「ポリ乳酸製架橋材の製造方法およびポリ乳酸架橋材」
特願 2006-213951 号

06PAT009 2-16
玉田、八木、長澤（原子力機構）金澤（住友電工ファインポリマー）
「ポリ乳酸弾性体」
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06PAT010 2-18
瀧上、天田（群馬県産業支援機関）、長澤、八木、玉田、笠井、吉井（原子力機構）
「カルボキシメチルセルロースゲルの製造方法」
特願 2006-250947 号
06PAT011 2-18
瀧上（群馬県産業支援機構）、長澤、八木、玉田（原子力機構）
「カルボキシメチルセルロースを主成分とするゲルの製造方法とそのゲル」
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06PAT012 2-19
八木、長澤、廣木、玉田（原子力機構）、Charito
t. Aranilla（フィリピン原子力研究所）
「多糖類を原料とするゲルの製造方法」
特願 2006-331679 号

06PAT013 2-20
笠井昇、吉井文男（原子力機構）、湯浅淳一（積水化学工業）
「ポリオレフィン系樹脂管の曲げ加工方法」
特願 2007-129566 号

06PAT014 3-52
鵜飼光子、下山雄平（北海道教育大学）
「放射照射食品の検査方法」
特願 2006-161019 号

06PAT015 3-53
藤里俊哉、山田、永石（原子力機構）、「石英、アルミナなどの耐強酸性の酸化物を触媒とする放射線誘起水素製造法」
特願 2007-154225 号

06PAT016 3-54
河地有木、藤巻秀、松橋信平（原子力機構）
「光合成機能イメージングのための新規方法」
特願 2007-64288 号

06PAT017 3-55
杉本雅樹、吉川正人（原子力機構）、関修平、佃諭志、田川精一（大阪大学）
「イオンビーム照射によるセラミックナノワイヤーの製造法」
特開 2007-76978 号

06PAT018 4-19
山田、永石（原子力機構）
「石英、アルミナなどの耐強酸性の酸化物固体を触媒とする放射線誘起水素製造法」
特願 2007-64288 号

06PAT019 4-19
鵜飼光子、下山雄平（北海道教育大学）、湯浅淳一（積水化学工業）
「光合成機能イメージングのための新規方法」
特願 2007-64288 号

06PAT020 4-19
河地有木、藤巻秀、松橋信平（原子力機構）
「光合成機能イメージングのための新規方法」
特願 2007-64288 号

06PAT021 4-25
関修平、佃諭志、田川精一（大阪大学）、杉本雅樹、吉川正人（原子力機構）
「高分子多段ナノワイヤー及びスターパースト型ナノ粒子とそれらの製造法」
特願 2007-64288 号

06PAT022 4-25
関修平、佃諭志、田川精一（大阪大学）、杉本雅樹、吉川正人（原子力機構）
「高分子多段ナノワイヤー及びスターパースト型ナノ粒子とそれらの製造法」
特願 2007-64288 号
06PAT023 4-49
島田、及川、加藤、佐藤、堀内、遊佐、岸、中野（群馬大学）、佐藤、酒井、上松、福田、荒川（原子力機構）
「荷電粒子線の照射位置決定装置、その使用方法、及び照射位置決定装置を用いた治療装置」
PCT/JP2007/050347

06PAT024 4-51
吉田、田島（原子力機構）、横山（信越化学）
「ECR イオン源に用いられるプラズマ閉じ込め用のミラー磁場発生装置及び方法」
特開 2006-049020 号
Appendix 3

List of Related Press-Release and TV Programs

06NP001  2-01
平成18年7月28日、日経産業新聞に掲載、
「ホタテ内臓 捨てずに利用 無害化し肥料などに」

06NP002  2-16
平成19年6月21日、25日、26日読売新聞他7紙に掲載、
「塩ビに替わる柔らかいポリ乳酸の開発に成功—植物由来プラスチックの応用範囲を大幅に拡大—」

06NP003  2-18
平成18年12月8、12、21日、日本経済新聞他8紙に掲載、
「環境に優しい植物由来のゴムのような弾性ゲル体の開発に成功」、「吸水・弾力性のゲル開発」

06NP004  3-01
平成18年4月25日、毎日新聞他8紙に掲載、
「植物を紫外線に強くするメカニズム」

06NP005  4-33
平成18年8月7日、上毛新聞他4紙に掲載、
「高輝度陽電子ビームを用いて表面ナノ物質の原子立体配列の観測に成功—陽電子で物質最表面の顕微技術が可能に—」

06NP006  4-54
平成19年2月20日、日刊工業新聞他7紙に掲載、
「原子力機構と芝浦工大PBWで長深度加工 神鋼と先端素子装置開発へ」「陽子ビーム加工技術開発 原研機構など」「プロトンビームライティング 超精密金型作製に応用 原子力機構—芝浦工大—神戸製鋼」等

06TV001  3-33
平成18年8月3日19時30分、NHK総合、[クローズアップ現代]、
「残留放射線の脅威 ～第3の被爆を追う～」
## Appendix 4 Type of Research Collaboration

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Type of Research Collaboration:
- Joint Res.: Joint research with private company or governmental institution
- Coop. Res. Univ.: Cooperative research with a university or universities
- Joint Res. Proj.: The Joint Research Project #

Irradiation Facilities:
- C: AVF Cyclotron System
- T: Tandem Electrostatic Accelerator
- I: Ion implanter
- S: Single-ended Electrostatic Accelerator
- G: Co-60 Gamma-Ray Irradiation Facilities
- E: Electron Accelerator

O.L.: Off-shore
Symbol used in the Appendix

Written example

<table>
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<th>173</th>
<th>4-42</th>
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</thead>
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① Abbreviation of A.D. year

② J : Publications in Journal
    C : Publications as Proceeding
    NP : Press-Release
    TV : TV Programs
    PA : Patent

③ Reference number

④ The number of research field

1  Space, nuclear and energy engineering
2  Environment conservation and resource security
3  Biotechnology and medical application
4  Advanced materials, analysis and novel technology

⑤ The number of papers

⑥ Irradiation Facilities

    C  AVF Cyclotron System
    T  Tandem Electrostatic Accelerator
    I  Ion implanter
    S  Single-ended Electrostatic Accelerator
    G  Co-60 Gamma-Ray Irradiation Facilities
    E  Electron Accelerator
    O  Off-line
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国際単位系（SI）

表1. SI基本単位

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表2. 基本単位を用いて定義されるSI組立単位の例

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<td>m/s</td>
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<td>キルワ</td>
<td>W</td>
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表3. 固有の名称とその独自の記号で定義されるSI組立単位

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<td>貧</td>
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<td>A</td>
<td>ルツル×秒の量</td>
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<td>G</td>
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<tr>
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表4. SI組立単位で定義される物質の単位

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表5. SI組立単位の例

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表6. 国際単位系と併用される国際単位系に属さない単位

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表7. 国際単位系と併用される国際単位系に属さない単位と併用されるその他の単位

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表8. SI組立単位で定義される物質の単位

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(第7版, 1998年改訂)