VIII-Ⅱ-1. Project Research

Project 11

PR11 Behavior of Radioactive Nuclides in Intense Radiation Fields in Accelerator Facilities

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OBJECTIVES AND RESEARCH SUBJECTS: Recently high-energy and high-intensity accelerators have been developed. They are widely used for many fields such as physics, chemistry and cancer therapy. The high-energy proton accelerators produce high-energy primary and secondary hadron particles, and a variety of radionuclides are formed through nuclear reactions including spallation reactions in high-energy accelerator facilities. The activation takes place in accelerator hardware, air, cooling water and other materials such as soil outside of shielding concrete walls.

From the viewpoints of radiation safety and safe accelerator operation, the management of the formed radionuclides is very important. The behavior and property of the radionuclides is the most important information to estimate radiation exposure and to ensure the accelerator safety.

In this work, the behavior and property was investigated in the air of the accelerator rooms and the cooling water. In addition, problems concerning activation in accelerator facilities were also investigated to obtain correct estimation of amount of the radionuclides formed in air and accelerator hardware.

The following problems are studied in this work.

1. The property of radioactive airborne species (radioactive aerosols and radioactive gases) in the air.
2. The behavior of dissolved metals from cooling water tubes and radioactive nuclides formed in the water.
3. Problems related to activation in accelerator facilities such as development of new radiation detectors for high energy accelerators and measurement of formation cross section of the principal radionuclides for dose estimation.

The participating research subjects (PRS) in Fiscal 2012 and co-workers in each subject are listed below.

PRS-1 (24P11-1) Difference of number and radioactivity size distribution of airborne aerosols formed in target room depending on beam currents
(S. Yokoyama, Y. Oki and S. Sekimoto)

PRS-2 (24P11-2) Measurement of Radioactive Airborne Nuclides in an FFAG Accelerator
(Y. Oki, K. Yamashita[1], Y. Ishi, T. Uesugi and Y. Kuriyama)

PRS-3 (24P11-3) Corrosion of Metals and Colloid Formation in Water under Intense Raiation Field
(K. Bessho, H. Matsumura, Y. Oki[1], N. Akimune[2], S. Sekimoto, M. Hagiwara, K. Iijima, T. Sanami, K. Masumoto, J. Hori, N. Abe)

MAIN RESULTS:

PRS-1

Radioactive size distribution and radioactivity of airborne aerosols formed on different beam current intensity in the target room of the electron linear accelerator in KURRI were estimated. The size distribution was measured with a scanning mobility particle sizer (SMPS) and a low pressure impactor. The size dependence on beam current was discussed for both of ^{13}N-bearing aerosol particles and non-radioactive aerosol particles.

PRS-2

An air irradiation experiment was performed for the first time in an FFAG proton accelerator in order to study the property of the radiation-induced radioactive airborne nuclides. The radioactivity and the number of the particles were measured. Preliminary results obtained in the first irradiation experiment were reported.

PRS-3

Metallic copper (Cu) contacted with water were irradiated with γ-rays in the ^{60}Co irradiation facility in KURRI, bremsstrahlung, or neutrons in the electron linac, as a model environment inside accelerator facilities. Radiation effects on corrosion of Cu in water and formation of colloidal Cu species were observed.

In addition, a new radiation monitor for burst neutron field was tested under a burst neutron field that was generated using a Ta target bombarded with electrons from the linac. The monitor has been developed to measure dose rate from beam loss of a pulse-mode accelerator to prevent activation accelerator hardware.

採択課題番号 24P11 加速器施設の高線量場における放射性核種の挙動に関する研究　プロジェクト（京大・原子炉）沖　雄一

PR11-1 Different of Number and Radioactivity Size Distribution of Airborne Aerosols Formed in Target Room Depending on Beam Currents

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INTRODUCTION: Radioactive aerosols are formed by beam irradiation around a beam line and in a target room of accelerator facilities. The inhalation of the radioactive aerosols may results in internal exposure for accelerator maintenance workers. The radioactive size distribution of aerosols is important factor to estimate internal dose. The time variations of number size distribution and formation mechanism of airborne aerosols formed in the accelerator facilities were investigated in the past studies [1-3]. In this study, radioactive size distribution and radioactivity of airborne aerosols formed on different beam current intensity in the target room were estimated.

EXPERIMENTS: In the target room of KUR-LINAC, a water cooled Ta target was irradiated with 30MeV electrons beam with currents of 20, 50, 80, 100 and 120μA for about 2 hours per experiment and neutrons were generated. The number-size distribution of airborne aerosols formed in the target room was measured continuously by SMPS (Model Series 3936, Tokyo Dylec corp.) during the experiments. The radioactivity-size distribution of aerosols was collected by a low pressure cascade impactor (Tokyo Dylec corp.) from 60 to 90 minutes after irradiation start. The radioactivity of aerosols collected on each stage of the cascade impactor were measured by an automatically α and β particles detector (JDC-5300, Hitachi Aloka Medical, Ltd). The aerosols were collected by filter and detected by BGO scintillation counter. As this result, it was found that ^{13}N made a large contribution to total activity. The radioactivity-size distribution of aerosols was also estimated by using the measured number-size distribution of aerosols and the attachment coefficient of ^{13}N.

RESULTS: As shown in Fig. 1, the number-size distribution of aerosols had two peaks. The second large peak was observed at 110-140nm diameters independently of the beam currents. The peak of measured activity size distribution of aerosol was also appeared at 230-260nm diameters regardless of the beam currents although the distribution had only a single peak (Fig. 2). Total measured radioactivity of aerosols was linearly proportional to beam current. It is considered that the growth of radioactive aerosols is avoided even if the amount of generated aerosols is large because the formed radioactive aerosols diffuse sufficiently in the target room. Fig. 3 shows measured and calculated activity size distribution of aerosols on beam currents of 20 and 100μA. The calculated distribution on the beam current of 20 and 100μA had peaks at 133 and 152 nm diameters, respectively. The diameter that the peak activity of calculated distribution appeared is smaller than the measured one.

ACKNOWLEDGMENTS: In these experiments, measurement of the number-size distribution of aerosol using SMPS system was supported by Tokyo Dylec corp.

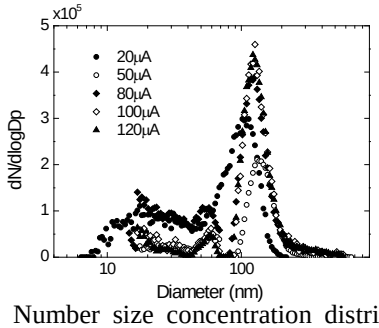

Fig. 1. Number size concentration distribution of airborne particles at beam currents of 20-120μA at 60minitues after the beam irradiation started.

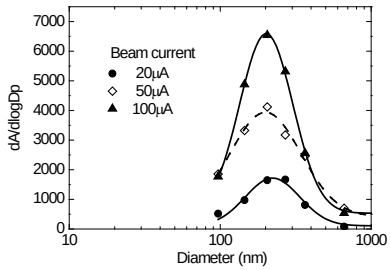

Fig. 2. Measured activity size distribution at beam currents of 20, 50 and 100μA from 60 to 90 minutes.

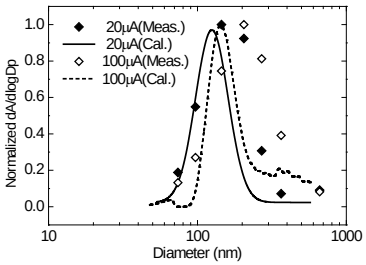

Fig. 3. Activity size distribution of ^{13}N aerosol measured and estimated at current in the case of the beam current of 20and 100μA.

REFERENCES:

[1] S. YOKOYAMA *et al.*, Jpn. J. Health. Phys., **43** (2008) 333-340.
[2] S. YOKOYAMA *et al.*, Rad. Prot. Dosi., 127 (2007) 392-397.
[3] N. Osada, Behavior and formation mechanism of radioactive aerosol in accelerator facilities (Kyoto University s) (2012).

PR11-2 Measurement of Radioactive Airborne Nuclides in an FFAG Accelerator

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INTRODUCTION: In accelerator facilities, radioactive airborne species are formed in air of the accelerator rooms during machine operation. Oxygene-15 (half life: 2 min), ^{13}N (10 min) and ^{11}C (20 min) are principal radionuclides in the air [1]. In addition, ^{7}Be (half life: 53 d) is also formed through nuclear spallation reactions from air in high-energy proton accelerators. The ^{7}Be atoms in air in the accelerator rooms are observed as aerosol particles; however, ^{15}O, ^{13}N and ^{11}C exist in both states of gas molecules and the aerosol particles. Their states are very important information on accelerator radiation safety management.

In this work, an air irradiation experiment was performed for the first time in an FFAG proton accelerator in order to study the property of the radiation-induced radioactive airborne nuclides. The radioactivity and the number of the particles were measured. Preliminary results obtained in the first irradiation experiment are described below.

EXPERIMENTS: The irradiation experiment was carried out in the 150-MeV FFAG (Fixed Field Alternating Gradient) Proton Accelerator of Research Reactor Institute, Kyoto University (KURRI). In the FFAG accelerator system, an 11-MeV H^- ion beam is injected to the FFAG main ring by the H^- linear accelerator. Protons are accelerated to 150 MeV after conversion of H^- ions to protons. The specimen irradiation beam line, one of the extraction beam lines of the main ring was used for air-irradiation.

The air-irradiation chamber was installed at the end of the specimen irradiation beam line before the beam dump. The chamber was a 1-m long stainless steel cylinder with the internal diameter of 15 cm. The beam pipe and the chamber were separated with a 1-mm thick Al plate. The aerosol-free air was introduced to the chamber at the flow rate of 20 L/min, and the irradiated air was sampled from the chamber. Fig. 1 shows the irradiation chamber together with the specimen irradiation beam line and the FFAG main ring.

The activity of the sampled air was measured with a 1.5-L ionization chamber after removing aerosol particles by filtration. The fluctuation of the beam intensity was monitored using this ionization chamber measurement. The radiation-induced aerosol particles in the air were collected on a PTFE membrane filter (pore size: 0.45-μm). The γ-spectra of the filters were measured with Ge detectors. In addition, the 511-keV annihilation γ-rays from the positron emitters were measured with a coincidence system of two bismuth germanate (BGO) scintillators.

The number concentration of the aerosol particles formed in the irradiated air was counted with a Condensation Particle Counter (CPC) (TSI Inc., Model 3025A).

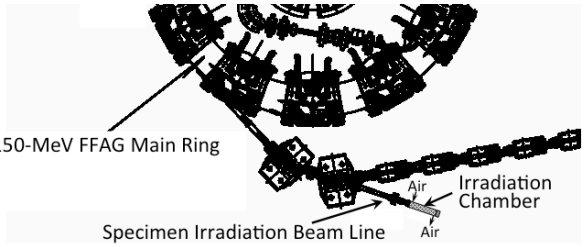

Fig. 1 Irradiation Chamber

RESULTS AND DISCUSSION: Dense aerosols are observed in the irradiated air. The number concentration exceeded the upper detection limit of the CPC (10^5 particles/cm^3). The 511-keV annihilation peak due to the positron emitters and the ^{7}Be peak of 478 keV were observed in the γ-spectrum of the collected aerosols. The decay curve of the annihilation peak was fitted to the principal positron emitters by a least-squares method. The radionuclide formed in air in the bremsstrahlung irradiation in the electron linac of KURRI was almost ^{13}N at the time of measurement [2]; however, ^{11}C was observed in addition to ^{15}O and ^{13}N in the proton irradiation. From the viewpoint of radiation safety, attention should be paid to the formation of ^{11}C whose half life is longer than other positron emitters.

Dense aerosol particles supplied at the constant flow rate are useful for calibration of size-measuring instruments for aerosol particles. Because stable production of fine particles (nano- and sub-micron particles) is more difficult than micron-order particles, devices that easily produce the fine particles can be employed for calibration of aerosol-measuring instruments for the fine particles. This air-irradiation system in the proton accelerator can be applied to the particle production device for the fine particle calibration by combining with a Differential Mobility Analyzer (DMA) that separates particles of desired size range from the radiation-induced particles.

REFERENCES:

[1] A. Endo, Y. Oki, Y. Kanda, T. Oishi, and K. Kondo, Radiat. Prot. Dosim., **93** (2001) 223-230.
[2] N. Osada, Y. Oki Y, K. Yamasaki and S. Shibata, Progress in Nuclear Science and Technology, **1** (2011) 483-486.

採択課題番号 24P11-2　加速器室の空気に生成する放射性化学種の生成挙動　プロジェクト
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（藤田保健衛生大医療科学部）横山須美

PR11-3 Corrosion of Metals and Colloid Formation in Water under Intense Radiation Field

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INTRODUCTION: At high-intensity accelerator facilities, intense radiations affect chemical states of elements inside the facilities. In the cooling-waters for various components, formation of metal related colloids in water is one of the important subjects in the radiation managements. In this work, metallic copper (Cu) contacted with water were irradiated by γ-rays, bremsstrahlung, or neutrons, as a model environment inside accelerator facilities. Radiation effects on corrosion of Cu in water and formation of colloidal Cu species were studied.

EXPERIMENTS: Cu vessels (*I. D.* 19 x 75 mm) filled with pure water were irradiated by γ-rays generated at the ^{60}Co γ-ray irradiation facility, or bremsstrahlung and neutrons generated by 30 MeV electron beam hitting on a Ta target (*Φ*50 x 62 mm) at the electron linear accelerator (LINAC). Samples at LINAC experiments were fixed at the downstream position (80 mm from Ta target) and perpendicular position (80 mm from Ta target).

After the irradiations, water samples in Cu vessels were poured out and treated with four kinds of ultrafiltration (UF) units for particle size separation. Estimated pore sizes of the UF units were 200, 16, 7 and 3 nm. Concentrations of Cu in the filtrates and unfiltered samples were determined by ICP-AES analyses.

RESULTS: Figure 1 (a) and (b) show soluble and colloidal concentrations of Cu in water as a function of reaction time. The data for LINAC experiments were obtained at the downstream sample position where flux of neutrons can be neglected. Therefore, Fig. 1 shows the effects of photon irradiation, and it demonstrates that irradiation of high-energy photons affects the corrosion of Cu and colloid formation in water phase.

Concentration of soluble Cu species increased at photon irradiations. In γ-ray irradiation at the dose rate of 2.5-2.9 kGy/h, soluble Cu concentration increased with reaction time and reached to a plateau concentration of 0.6 mg/L. At the dose rate of 19-22 kGy/h, a plateau concentration of soluble Cu was 1.7 mg/L. In the LINAC experiments, soluble Cu concentration also increased with irradiation times. These results imply that dissolution of Cu progresses under intense photon field and finally reaches to plateau concentrations dependent on the photon intensity and/or energy.

Formation of colloidal Cu species was also noticeable under intense photon field. In contrast with soluble fractions, colloidal Cu concentration quickly reached to plateau within 2 h. More comprehensive analyses and discussion are in progress.

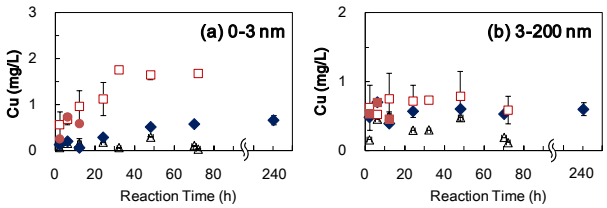

Fig. 1 Cu concentrations in water in the copper vessels after the irradiations as a function of reaction time.
(a) Soluble fraction:0-3 nm; (b) Colloidal fraction:3-200 nm; Symbols △ :without irradiation; ◆ :γ-ray (2.5-2.9 kGy/h); □:γ-ray (19-22 kGy/h);●: LINAC (current:10 μA)

RADIATION MONITOR FOR BURST NEUTRONS: A radiation monitor for burst neutron field has been developed to measure dose rate from beam loss of a pulse-mode accelerator [1]. The monitor, which consists of a ^{3}He proportional counter, a 6.5 cm thick polyethylene moderator, equips a newly developed charge integrate circuit (current readout). The monitor was tested under a burst neutron field that was generated using a Ta target bombarded with electrons from LINAC, at 120 degree, 10 m from the target. Another monitor with a conventional pulse readout circuit and the same moderator also tested for comparison. Photon dose of this field was measured using an ionization chamber. The intensities of the burst field were varied with changing the beam energy and pulse width. Fig. 2 shows experimental results on measured dose rate as a function of number of neutrons generated at the target. The current readout type neutron monitor showed linear response up to neutron dose rate of 400 μSv/h. The photon dose rate that is four times higher than neutron one, has less impact on the neutron monitor because of photon sensitivity.[1] The pulse readout monitor failed at low dose rate owing to serious pulse pile-up.

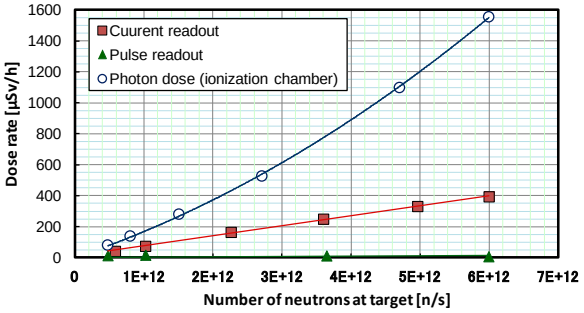

Fig. 2 Comparison of the dose rate measured with the different readout type neutron monitors. Photon dose was measured using ionization chamber.

REFERENCES:
[1] K. Iijima *et al.*, Prog. Nucl. Sci. Tech., 1 (2011) 300

採択課題番号 24P11-3　高線量放射線場における水中金属材料の腐食およびコロイド生成挙動の解析　　プロジェクト

（KEK・放射線）別所光太郎、松村宏、桝本和義、萩原雅之、飯島和彦、佐波俊哉
（京大・原子炉）沖雄一、関本俊　　（京大院・工）秋宗尚弥