PR5 Behavior of Radioactive Nuclides in Intense Radiation Fields in Accelerator Facilities

Y. Oki

Research Reactor Institute, Kyoto University

OBJECTIVES AND RESEARCH SUBJECTS:

High-energy and high-intensity accelerators are widely used for many fields such as physics, chemistry and cancer therapy. The high-energy proton accelerators produce a variety of radionuclides 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 radionuclides is very important. The behavior and property of the radionuclides are the most important information to estimate radiation exposure and to ensure the accelerator safety.

In this work, the behavior and property were investigated in the air of the accelerator rooms and the cooling water.

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 the final year of this project research (Fiscal 2014) and co-workers in each subject are listed below.

PRS-1 (26P5-1) Radioactivity Size Distribution of Airborne Aerosols Formed in Target Room Estimated from Number Size Distribution
(S. Yokoyama, Y. Oki and S. Sekimoto)

PRS-2 (26P5-2) Production of Airborne Nano-Particles by Proton Irradiation in an FFAG Accelerator
(Y. Oki, N. Osada, S. Yokoyama, Y. Ishi, T. Uesugi, Y. Kuriyama and M. Sakamoto)

PRS-3 (26P5-3) Corrosion of Various Metals in Water and Formation of Metal-related Colloidal Particles under Intense Photon-Radiation Field
(K. Bessho, H. Matsumura, Y. Oki, S. Sekimoto, K. Masumoto and N. Abe)

MAIN RESULTS:

PRS-1

The present study analyzed a distribution of radioactive aerosols in the target room of the KUR-LINAC. The size and radioactivity of the aerosol sampled from two sampling positions (40 cm and 230 cm apart from the Ta target) were compared. The target was irradiated with the 30-MeV electron beams of 10, 50, and 100-μA currents for ~2 h per experiment. Radioactive aerosols were collected with a low-pressure cascade impactor to obtain the particle size distribution. Under 10 μA and 50 μm, the particle size distribution for radioactive aerosols showed no significant difference between the two sampling positions. Total radioactivity at both positions increased with beam current.

PRS-2

An air irradiation experiment was performed in the 150-MeV FFAG proton accelerator using an air-irradiation chamber under various beam conditions. The size and concentration of the aerosol particles produced in the irradiation were studied for applications to the instrument calibration and other experiments using standard aerosol particles. The size was increased as the air supplying flow rate to the chamber was decreased. In other words, the particles grew as the irradiation time was increased. The size was possible to be varied by changing the flow rate to the chamber. In the case of 1 nA, beam current, the size was able to be changed from approximately 10 to 20 nm in geometric mean diameter. The size and concentration were also dependent on the beam current. An increase in size and concentration was observed above the proton current of approximately 300 pA and they were almost saturated above 1 nA at the flow rate of 18 L/min

PRS-3

In this work, experimental metal vessels filled with pure water were irradiated by γ-rays or bremsstrahlung / neutrons generated at electron linear accelerator (LINAC) facility as model environments for cooling water used inside accelerator facilities. Radiation effects on corrosion of various metals in water and colloid/particle formation of metal elements were systematically studied. After the irradiation of γ-rays or bremsstrahlung, soluble (<3 nm), colloidal (3-200 nm), and particulate (>200 nm) metal elements were detected in water. The size profiles for each element in water and characteristics in radiation effects were dependent on vessel materials. In the experiments using Al and Fe vessels, total Al/Fe concentrations became higher dependent on photon irradiation intensity. In the water supernatant obtained after the irradiations, Al and Fe were found to be mainly present as relatively-large particles (>200 nm). Concentrations of soluble species and fine colloidal species are quite small compared to those of the large particle species. Their chemical states were are also discussed in this work.

採択課題番号 26P5　　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

PR5-1

Radioactivity Size Distribution of Airborne Aerosols Formed in Target Room Estimated from Number Size Distribution

S. Yokoyama,[1] Y. Oki,[2] and S. Sekimoto[2]

[1] *School of Health Science, Fujita Health University*
[2] *Graduate School of Engineering, Kyoto University*

INTRODUCTION: To assess internal exposure of accelerator facility workers due to inhaling radioactive aerosols and gases, the radioactivity size distribution and the amount of radioactive aerosols formed in the facilities must be estimated [1–4]. In previous studies, aerosol radioactivity and number size distributions were measured in the target room of KUR-LINAC and the radioactivity size distributions were estimated from the number-size distributions [3, 4]. In addition, the relationship between radioactivity size distribution of aerosols and beam currents was investigated. The present study analyzes a distribution of radioactive aerosols in a target room.

EXPERIMENTS: Two sampling ports were prepared 40 cm (port 1) and 230 cm (port 2, near the wall surface) from the target of KUR-LINAC. Electron beams were generated under a 30-MeV voltage and 10-, 50-, and 100-μA currents for ~2 h per experiment. A water-cooled Ta target was placed at the end of the beam line as a neutron generator. Radioactive aerosols were collected by a 13-stage low-pressure cascade impactor (Tokyo Dylec Corp.). Stainless steel plates coated with grease were set in each stage, except for the lowest stage, in which a filter was set. The collection started at 60 min after irradiation began and continued for 30 min. After collection, the plate and filter radioactivities were measured by a GM survey meter (TGS123-C, Aloka). Radionuclides formed in the target room confirmed that ^{13}N contributed significantly to total activity, when compared with the lower radioactivity measured in the previous study [4].

RESULTS: Figure 1 shows the radioactivity size distribution of aerosols under 10- and 50-μA beam currents at ports 1 and 2. Under 10 μA, the median diameters of radioactive aerosols were 270 and 189 nm (σ: 1.5 and 1.2) at ports 1 and 2, respectively. Under 50 μA, median diameters of radioactive aerosols were 270 and 229 nm (σ: 1.6 and 1.4) at ports 1 and 2, respectively. In the previous study, the median diameter of radioactive aerosols collected near the target was 261±3.9 nm under 20-100 μA [3, 4]. Figure 2 illustrates the total radioactivity of aerosols under 10, 50 and 100 μA. Total radioactivity at ports 1 and 2 increased with beam current.

Radionuclides are mainly generated by interaction between neutrons and the atmospheric nitrogen and oxygen proximal to the target just after the electron beam hits the target. Non-radioactive aerosols were formed by irradiation by electrons leaked from the beam line and gamma rays induced by electrons.

Under 50 μA, the peak radioactivity count collected at port 2 was 1.3 times higher than that at port 1, as shown in Fig. 1. The number size distributions of aerosols simultaneously measured by a Scanning Mobility Particle Sizer (TSI, Model 3080) showed a similar result. Radioactive aerosols are formed by deposition of radionuclides on non-radioactive aerosols. Thus, the number of radioactive aerosols at port 2 under 50 μA should increase. No clear difference existed between median diameters of radioactive size distributions at ports 1 and 2. This means that the non-radioactive aerosols were generated not only around the target, but also throughout the target room.

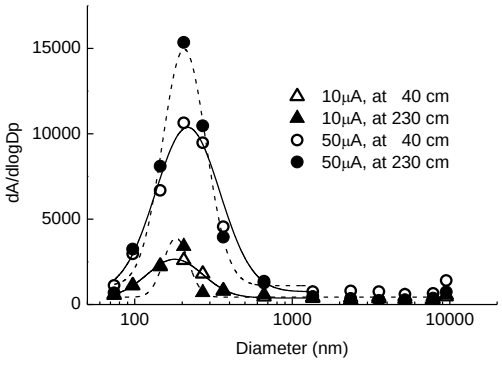

Fig. 1. Activity size distribution of radioactive aerosols collected at ports 1 and 2.

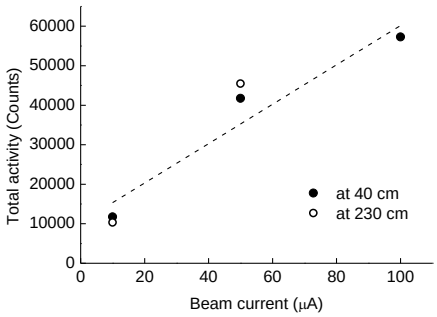

Fig. 2. Total activity of radioactive aerosols at ports 1 and 2.

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] S. YOKOYAMA et al., KURRI progress report 2012, PR11-1 (2013), http://www.rri.kyoto-u.ac.jp/JRS/.
[4] S. YOKOYAMA *et al.*, KURRI progress report 2013, http://www.rri.kyoto-u.ac.jp/PUB/report/PR/ProgRep2013/Project8.pdf

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採択課題番号 26P5-1　　加速器施設内生成粒子の粒径分布に関する研究　　プロジェクト

（藤田保衛大・医科）横山　須美（京大・原子炉）沖　雄一、関本　俊

PR5-2 Production of Airborne Nano-Particles by Proton Irradiation in an FFAG Accelerator

Y. Oki, N. Osada[1], S. Yokoyama[2], Y. Ishi, T. Uesugi, Y. Kuriyama and M. Sakamoto

Research Reactor Institute, Kyoto University
[1]Advanced Science Research Center, Okayama University
[2]School of Health Science, Fujita Health University

INTRODUCTION: In accelerator facilities, very fine radiation-induced aerosol particles were often produced in air of accelerator rooms during machine operation. The size and concentration of the particles are generally very stable under a given beam condition. This particle generation may be applicable to calibration of size-measuring instrument for the fine particles in the diameter range of nm to 0.1 μm.

In this work, an air irradiation experiment was performed in an FFAG (Fixed Field Alternating Gradient) proton accelerator under various beam conditions. The size and concentration of the aerosol particles produced in the irradiation were studied for applications to the instrument calibration and other experiments using standard aerosol particles.

EXPERIMENTS: The irradiation experiment was performed in the 150-MeV FFAG proton accelerator of Research Reactor Institute, Kyoto University (KURRI). The FFAG accelerator system consists of the 11-MeV H^- linear accelerator (Injector) and the FFAG main ring. The injected protons are accelerated in the main ring to 150 MeV after conversion of H^- ions to protons. The nominal 150-MeV proton beams were extracted to an air irradiation chamber in the specimen irradiation beam line. Figure 1 shows the irradiation chamber together with the specimen irradiation beam line and the main ring.

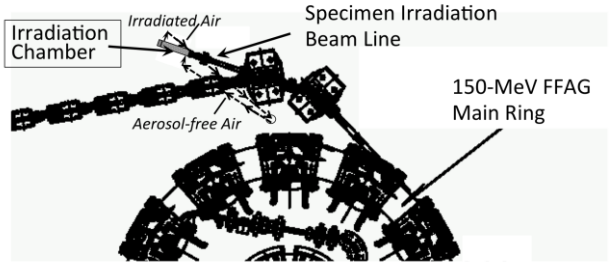

Fig.1 Irradiation Chamber

The air-irradiation chamber was a 1-m long stainless steel cylinder (i.d. 15 cm). It was installed upstream of the beam dump of the specimen irradiation beam line. The 1-mm thick aluminum plates were used as both ends of the chamber. The aerosol-free air was prepared by filtration and was introduced to the chamber at the constant flow rate, and the irradiated air was sampled from the chamber at the same flow rate.

The size distribution and the total concentration of the aerosol particles were measured with SMPS (Scanning Mobility Particle Sizer).

RESULTS AND DISCUSSION: The size of formed aerosol particles showed very steady mono-modal log-normal distributions. The size was increased as the air supplying flow rate was decreased. In other words, the particles grew as the irradiation time was increased. The size was possible to be varied by changing the flow rate to the chamber. In the case of 1 nA, beam current, the size was able to be changed from approximately 10 nm (the flow rate: 18 L/min) to 20 nm (8 L/min) in geometric mean diameter. The size and concentration were also dependent on the beam current. An increase in size and concentration was observed above the proton current of approximately 300 pA and they were almost saturated above 1 nA at the flow rate of 18 L/min [1].

During the air irradiation, principal radionuclides formed in the accelerator air, ^{15}O (half life: 2 min), ^{13}N (10 min), ^{11}C (20 min) [2] and ^{7}Be (53 d), are formed together with the radiation-induced aerosol particles. The aerosol particles form radioactive aerosols by incorporating the radioactive atoms into the radiation-induced particles. The size distribution of ^{7}Be-bearing aerosol particles were successfully measured with a graded screen array system in the same air-irradiation chamber [1]. The ^{7}Be aerosol particles can be also used in various research including calibration of aerosol instruments for radioactive fine particles.

In conclusion, the air-irradiation chamber was capable of producing aerosol particles in the diameter range of 10 to 20 nm by the 1-nA proton irradiation carried out in the FFAG accelerator.

REFERENCES:

[1] Y. Oki, N. Osada, Y. Ishi, T. Uesugi, Y. Kuriyama and M. Sakamoto, KEK Proceedings **2014-7**, 52-57.
[2] A. Endo, Y. Oki, Y. Kanda, T. Oishi, and K. Kondo, Radiat. Prot. Dosim., **93** (2001) 223-230.

採択課題番号 26P5-2　　加速器室の空気生成する放射性化学種の生成挙動　　プロジェクト
（京大・原子炉）沖　雄一、関本俊、（藤田保健衛生大医療科学部）横山須美

PR5-3 Corrosion of Various Metals in Water and Formation of Metal-related Colloidal Particles under Intense Photon-Raiation Field

K. Bessho, H. Matsumura, Y. Oki[1], S. Sekimoto[1],
K. Masumoto, N. Abe[1]

Radiation Science Center, KEK
[1]Research Reactor Institute, Kyoto University

INTRODUCTION: At high-energy accelerator facilities, intense radiations affect chemical states of elements including radionuclides inside the facilities. In the cooling-waters for various accelerator components, corrosion of metal components and formation of metal related colloids/particles in water are important subjects in the radiation safety managements at accelerator facilities, because some radionuclides are incorporated in colloidal species and it complicate the behavior of nuclides [1,2].

In this work, experimental metal vessels filled with pure water were irradiated by γ-rays or bremsstrahlung / neutrons generated at electron linear accelerator (LINAC) facility as model environments for cooling water used inside accelerator facilities. Radiation effects on corrosion of various metals in water and colloid/particle formation of metal elements were systematically studied.

EXPERIMENTS: Metal 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 / neutrons generated by 30 MeV electron beam hitting on a Ta target (*Φ*50 x 62 mm) at the LINAC facility. Samples at the LINAC experiments were placed at the downstream position (40 or 80 mm from Ta target) and perpendicular position (40 or 80 mm from Ta target). Monte Carlo calculations and activation experiments using metal foils (Au, Al) demonstrated that radiation effects observed in the present LINAC experiments could be ascribed to the effects of photon irradiations (maximum fluxes; 3 MeV for 0-deg and 0.7 MeV for 90-deg). Contribution of neutrons was found to be much smaller than those of photons in these experiments.

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

RESULTS: After the irradiation of γ-rays or bremsstrahlung, soluble (<3 nm), colloidal (3-200nm), and particluate (>200nm) metal elements were detected in water. The size profiles for each elements in water and characteristics in radiation effects were dependent on vessel materials (pure Fe, Al, Cu, and SUS-304, A6061 alloys). In this report, some characteristics observed for pure Al, Fe and Cu are described.

In the experiments using Al and Fe vessels, irradiations were carried out at LINAC with two kinds of beam currents (20, 100 μA) and the irradiation times of 2 h. Total Al/Fe concentrations became higher dependent on photon irradiation intensity. In the water supernatant obtained after the irradiations, Al and Fe were found to be mainly present as relatively-large particles (>200 nm). Concentrations of soluble species and fine colloidal species are quite small compared to those of the large particle species. Al and Fe in non-degassed water are considered to be in the oxidation states of III (Al^{3+}, Fe^{3+}), which tend to form hydroxide precipitations in neutral pH conditions. It can be supposed that photon irradiation facilitates the corrosion (oxidation) of pure Al and Fe metals in water, consequently Al and Fe are released as Al^{3+}, Fe^{3+} ions from the metal surface to water, finally form $Al(OH)_3$ and $Fe(OH)_3$ particles in water phase.

In case of the experiments using Cu metal vessels, effects of radiation were mainly observed as increase in soluble Cu species (<3 nm) and production of large particle Cu species (>200 nm). Under irradiation conditions, soluble Cu concentration increased with time depend on photon doses and finally reached to plateau, which may be originated from hydrolysis of Cu(II) ions. On the other hand, production of particle Cu species was observed remarkably after long irradiation times (> 20-30 h). These characteristic time-dependence imply that particle Cu species were initially generated at the Cu/water interface as particle-like corrosion products. These products grew up at the interface in a time scale of several tens hours, and were finally released into water phase as particles (> 200 nm). The particle production processes at the interface were also promoted by photon irradiations [3].

Radiation effects on corrosions of metals in water are explained by production of oxidative reactive species, such as OH radicals and H_2O_2, induced by radiolysis of water molecules. By using water quality testing kits, it was demonstrated that several mg/L of H_2O_2 and O_3 were produced by irradiation of water by γ-rays or bremsstrahlung, and these oxidative species should induce various radiation effects clarified by this work.

REFERENCES:

[1] K. Bessho *et al.*, J. Radioanal. Nucl. Chem., **303** (2015) 1719-1725.
[2] H. Matsumura *et al.*, Progress in Nucl. Sci. Technol., **4** (2014) 372-375.
[3] K. Bessho *et al.*, J. Radioanal. Nucl. Chem., **303** (2015) 1117-1121.

採択課題番号 26P5-3　高線量放射線場における水中金属材料の腐食および　プロジェクト
コロイド生成挙動の解析２

（KEK・放射線）別所光太郎、松村宏、桝本和義　（京大・原子炉）沖雄一、関本俊