VIII-Ⅱ-1. Project Research

Project 8

PR8 Behavior of Radioactive Nuclides in Intense Radiation Fields in Accelerator Facilities

Y. Oki

Research Reactor Institute, Kyoto University

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 2013 and co-workers in each subject are listed below.

PRS-1 (25P8-1) Estimation of the Radioactivity-Size Distribution of Airborne Aerosols Formed in the Target Room from the Number-Size Distribution
(S. Yokoyama, Y. Oki and S. Sekimoto)

PRS-2 (25P8-2) Measurement of Aerosol Particle Size in an FFAG Accelerator Using Wire Screens
(Y. Oki, Y. Ishi, T. Uesugi, Y. Kuriyama and M. Sakamoto)

PRS-3 (25P8-3) Corrosion of Copper and Colloid Formation in Water under Intense Photon-Radiation Field
(K. Bessho, H. Matsumura, Y. Oki, S. Sekimoto, K. Masumoto and N. Abe)

MAIN RESULTS:

PRS-1

Radioactivity size distribution of aerosols was measured with a low-pressure cascade impactor in the target room of the electron LINAC in KURRI. The aerosols were formed by the bombardment of the Ta target with a 30-MeV electron beam. The radioactivity size distribution was larger than the calculated size distribution. The impactor result is considered to be influenced by the deposition of radioactive gas on collection stages.

In addition, radioactivity was collected on the charcoal and the PTFE filters in the target rooms of the LINAC and FFAG facilities. The radioactivity of the charcoal filters was 100 times higher than that of the PTFE filters.

PRS-2

A new wire screen device for size measurement of very fine particles was assembled and examined using the FFAG proton accelerator. An air chamber installed in the FFAG main ring was irradiated with 150-MeV protons to produce radiation-induced radioactive aerosol particles in the range of nm to several tens of nm. The number size distribution obtained with the screen device was very similar to that obtained with SMPS. The activity size distribution for ^{7}Be was also obtained with the screen device. It was slightly larger than the number size distribution obtained with the screen device.

PRS-3

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

Corrosion mechanism enhanced by radiation was discussed based on the plots of the Cu concentration in the three size fractions vs. irradiation time.

採択課題番号 25P8　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

PR8-1 Estimation of the Radioactivity-Size Distribution of Airborne Aerosols Formed in the Target Room from the Number-Size Distribution

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

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

INTRODUCTION: To assess the internal exposure of radiation workers in accelerator facilities, it is important to estimate radioactivity size distribution and the amount of aerosols formed by beam irradiation around a beam line in a target room [1–3]. In a previous study, the measured radioactivity size distributions of aerosols did not agree with those estimated using the measured number–size distributions of aerosols and their attachment coefficients [3]. In the present study, the factors affecting the measured radioactivity-size distributions of aerosols were analyzed. In addition, the ratio of the amounts of the formed radioactive aerosols and gases, which is necessary to assess the internal exposure of the workers and clarify the formation mechanisms of radioactive aerosols, was estimated.

EXPERIMENTS: The experiments were conducted using the Kyoto University Reactor linear accelerator (KUR-LINAC) and the fixed-field alternating gradient (FFAG) accelerator facilities. In the LINAC, the 30-MeV electron beam was used to irradiate a water-cooled Ta target. The beam current was set to 40, 80 and 100 μA, with an irradiation period of ~2 h per experiment. In the FFAG, the 150-MeV proton beam current was set to 1 nA with an incidence period of 20 min.

In this experiment, the number–size distribution of aerosols formed in the target room was not measured. Thus, the radioactivity size distribution of aerosols was estimated using the number–size distribution measured under the same conditions as a pervious study [3]. The radioactive aerosols were collected on stainless steel plates set in a low-pressure cascade impactor (Tokyo Dylec Corp.). The collection started more than 30 min after irradiation. The radioactive aerosols and activation gases related to the aerosol formation were collected by PTEF filters (T080A047A, ADVANTEC) and charcoal filters (C248TA, ADVANTEC), respectively. For the charcoal filters, three sheets were overlapped. The radioactivity of each stage of the cascade impactor and the filters were measured by a GM survey meter (TGS123-C, Aloka) and a portable NaI (Tl) spectrometer (S-2361-88208, OKEN). It was confirmed by a measured decrease of radioactivity that ^{13}N made a large contribution to total activity.

RESULTS: Figure 1 shows the measured and calculated median diameter of radioactive aerosols in the target room of the LINAC. In the previous study, the median diameters calculated on the basis of number–size distribution of aerosols were about 60% of the measured ones. As a result of measuring the radioactivity, some radioactive nuclides were detected from the upper-stage plates on which the large particles were collected. However, particles of >1 μm diameter were not be formed in a previous study [1]. This might have been because the formed radioactive gases or small-size aerosols were deposited on the plates in the upper stages of the cascade impactor on which the large particles were collected. Thus the measured and calculated median diameters were compared for the particles of <0.6 μm. As a result, the difference between the calculated and measured diameters was <10% as shown in Fig. 1.

In addition, Table 1 shows the ratio of radioactivity of the charcoal and PTEF filters in the target rooms of the LINAC and FFAG facilities. The radioactivity of the charcoal filters was 100 times higher than that of the PTEF filters.

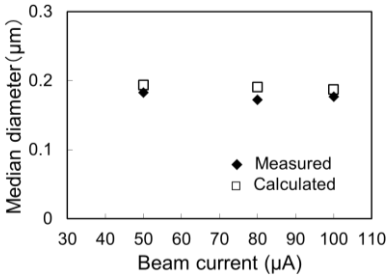

Fig. 1. Comparison between measured and calculated median diameters of radioactive aerosols under an electron beam energy 30 MeV in the LINAC.

Table 1. Ratio of radioactive aerosols and gases collected on the charcoal and PTEF filters in the target rooms of the LINAC and FFAG facilities.

Facilities	Beam energy and current	Ratio (%)	
		Charcoal filter	PTEF filter
LINAC	30 MeV, 40 μA	98.9	1.1
	30 MeV,100 μA	99.1	0.9
FFAG	150 MeV, 1 nA	99.8	0.2

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/.

採択課題番号 25P8-1　　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

PR8-2 Measurement of Aerosol Particle Size in an FFAG Accelerator Using Wire Screens

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

Research Reactor Institute, Kyoto University
[1]Graduate School of engineering, Tohoku University.

INTRODUCTION: In accelerator facilities, radioactive gas and radioactive fine aerosol particles are formed in air of the accelerator rooms during machine operation. The size of these airborne radioactive particles is very important information to estimate internal radiation dose; however, its measuring methods are very limited. Recently SMPS (Scanning Mobility Particle Sizer) has been widely used for size distribution measurement in the range of nm to 10 μm. However, the SMPS cannot be applicable to measurement of radioactivity-based particle size distribution. Although wire screen devices, such as graded screen array (GSA) and screen-type diffusion battery (SDB), are able to be used for both non-radioactive and radioactive particles, their accuracy is generally not better than SMPS.

In this work, a wire screen device (an improved GSA) was newly assembled, and was applied to size measurement for fine particles in the diameter range of 10 nm to several tens nm. The wire screen device was examined using a radioactive aerosol produced in an FFAG proton accelerator.

EXPERIMENTS: An air irradiation chamber was used for aerosol production by proton irradiation in the 150-MeV FFAG (Fixed Field Alternating Gradient) proton accelerator of Research Reactor Institute, Kyoto University (KURRI). The 18-L chamber was installed at the end of the specimen irradiation beam line of the FFAG main ring (Fig.1). Using flexible stainless steel tubes, aerosol-free air was supplied to the chamber from a measurement station and the irradiated air was returned to the station. The station was located in the room next to the accelerator room.

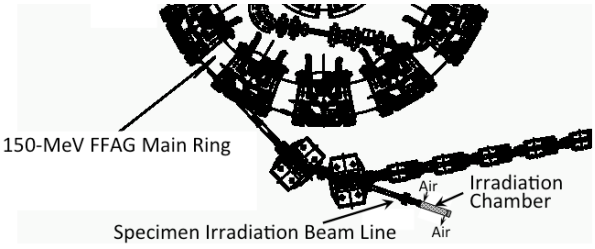

Fig.1 Air Irradiation Chamber

The FFAG accelerator supplied a stable 1-nA proton beam to the air irradiation chamber. At the measurement station the irradiated air was introduced to the wire screen device. The device consisted of a stack of wire screens and a back-up membrane filter. The stack comprised single 100, 200, 300-mesh screens and 12 500-mesh screens.

The air was sampled both from upstream and from downstream of the stack of the screens, and aerosol number concentration in the air was measured with a condensation particle counter (CPC). The number ratio of particles penetrating the screens (downstream of the screens) to whole particles before penetrating the screens is governed by their diffusion coefficient expressed by a function of particle size. The size distribution was calculated by fitting the curve of the penetrating ratios vs. the number of screens to the theoretical equation [1]. Log-normal distributions are assumed as the size distribution function in the calculation. For comparison the size measurement with SMPS was also carried out. The size of particles bearing a ^{7}Be atom was also analyzed by replacing the number concentration by the radioactivity of ^{7}Be measured with a Ge detector.

RESULTS AND DISCUSSION: Figure 2 shows the fitting results in the wire screen measurement carried out at the air sampling rate of 8 L/min, together with the size distribution obtained with SMPS at the same time. In the wire screen measurement the geometric mean diameter (d_g) was found to be 17.6 nm for whole particles, while 20.2 nm was obtained with SMPS. It was confirmed that the wire screen device correctly works in the fine particle measurement because the difference lies in the range of fluctuation during the sampling of the irradiated air from the chamber.

In the wire screen measurement, the size distribution of the ^{7}Be-bearing particles was slightly larger than that for whole particles. It may be attributed to attachment of ^{7}Be on the surface of an aerosol particle.

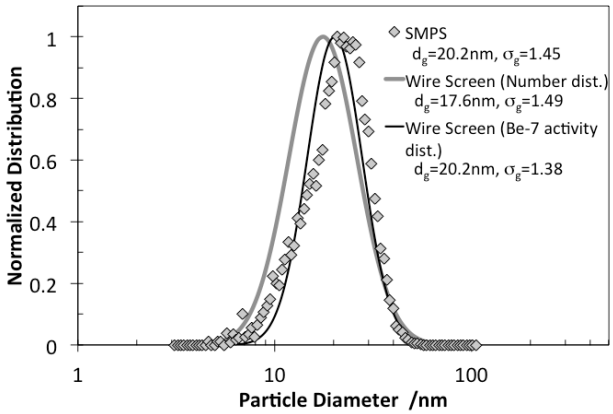

Fig.2 Comparison of particle size distributions obtained with GSA and SMPS

REFERENCES:

[1] Y.S. Cheng and H.C. Yeh, J. Aerosol Sci., **11** (1980) 313-320.

採択課題番号 25P8-2　　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
（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

PR8-3

Corrosion of Copper and Colloid Formation in Water under Intense Photon-Raiation Field

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

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

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 an important subjects in the radiation managements, because some radionuclides are incorporated in these colloidal species. In this work, metallic copper (Cu) contacted with water were irradiated by γ-rays or bremsstrahlung 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 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), (b) and (c) show soluble, colloidal, and particulate concentrations of Cu in water as a function of reaction time. These plots clearly shows the characteristic time-dependences for each size fractions.

Concentration of soluble Cu species (Fig. 1 (a)) increased with time under photon irradiation conditions. Increasing rates were remarkably high at the intense photon irradiation conditions; 0-deg position at LINAC (~12 kGy/h) and the source center in γ-ray irradiations (17-22 kGy/h). Under these conditions, soluble Cu concentration increased quickly to approximately 2 mg/L within reaction time of 20-30 h, and did not change significantly (or decreased slightly) thereafter. The Cu concentration of 2 mg/L correspond to the maximum soluble concentration of Cu(II) in water at neutral pH conditions, estimated from the solubility product constant for $Cu(OH)_2$. Thus it can be considered that apparent plateau concentration of Cu was originated from the hydrolysis of Cu(II) in water. After Cu(II) concentration reaches to the maximum solubility, excess Cu(II) ions form $Cu(OH)_2$ colloids/particles in water phase.

Concentration of nm-sized Cu-related colloids did not change significantly with reaction time (Fig. 1 (b)). On the other hand as shown in Fig. 1 (c), large particulate Cu species (>200 nm) began to increase in the reaction time of 20-30 h at two kinds of intense photon irradiation conditions. Under these intense radiation environments, the particulate Cu concentrations reached to the extremely high concentrations compared to other size fractions; 19 mg/L for irradiation at LINAC 0-deg position for 48 h ; 12 mg/L for irradiation of γ-ray (19 kGy/h) for 72 h.

It could be supposed that particle-like corrosion products, which may be copper oxides, were initially generated at the Cu/water interface. These corrosion products grew up at the Cu/water interface in several-tens hours. These processes may be enhanced under photon irradiation conditions, in which oxidative reactive species, such as H_2O_2, promotes corrosion of Cu surface more effectively. The particle-like corrosion products should be finally released into water phase as relatively-large particles (>200 nm), and these processes were observed as sudden increase of particulate Cu concentration in the reaction time of 20-30 h, as shown in Fig. 1 (c).

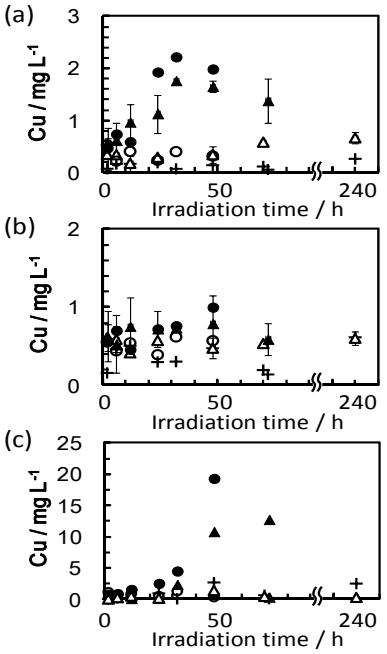

Fig. 1. Time-dependences of (a) soluble (0-3 nm), (b) colloidal (3-200 nm), and (c) particulate (>200 nm) concentration of Cu in water without irradiation, after the irradiation at 60Co γ-ray irradiation facility, and after the irradiation at electron LINAC.

Symbols; +:Without irradiation, ○:After the irradiation at electron LINAC (90-deg position), ●:After the irradiation at electron LINAC (0-deg position), △:After the irradiation of γ-ray from ^{60}Co source (2.3-2.9 kGy/h), ▲:After the irradiation of γ-ray from ^{60}Co source (17-22 kGy/h).

REFERENCES:

[1] K. Bessho *et al.*, J. Radioanal. Nucl. Chem., accepted

採択課題番号 25P8-3　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２　プロジェクト

（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