VIII-Ⅱ-1.　Project Research

Project 5

PR5

Studies on Radiation Safety Control at Accelerator Facilities

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Objectives and Allotted Research Subjects

A Fixed-Field Alternating-Gradient (FFAG) proton synchrotron (max energy 150 MeV, max mean current 1 nA) has been constructed next to the Kyoto University Critical Assembly (KUCA) to study the technical development for the accelerator driven subcritical reactor (ADSR). The proton beam was led to the A-core of KUCA to conduct the experiment of ADSR on March 4, 2009. On the other hand, the commissioning of a 30 MeV proton cyclotron was completed at the end of March, 2009, for the boron neutron capture therapy (BNCT). And other high energy accelerators are scheduled to conduct as neutron sources in the master plan of KURRI. In these situations, the radiation safety control at accelerator facilities is a key issue in the workplaces and environment.

In these projective studies, five research subjects were carried out in three facilities (the electron linear accelerator (LINAC), the Co-60 gamma irradiation facility and the proton cyclotron) as substitutes related to the accelerator fields.

The allotted research subjects (ARS) are as follows:

ARS-1 (23P5-1): Development of Passive Neutron Dosimeter with Good Energy Response.
(T. Iimoto, K. Shimada, K. Tani, Y. Fujimichi, K. Yamasaki and T. Takahashi)

ARS-2 (23P5-2): Production and Behavior of Tritium in Accelerator Laboratory.
(M. Ohta, S. Kimura, S. Fukutani and K. Okamoto)

ARS-3 (23P5-3): Corrosion of Metals and Colloid Formation in Water under Intense Radiation Field.
(K. Bessho, H. Matsumura, K. Masumoto, Y. Oki, N. Akimune, S.Sekimoto,K. Yamasaki and N. Osada)

ARS-4 (23P5-4):Adsorption of Radioactive Gas on a Membrane Filter in an Electron Linear Accelerator.
(Y. Oki, N. Osada and K. Yamasaki)

ARS-5 (23P5-5): Time Dependencies of Size Distributions of Radioactive Aerosols Produced in Accelerators.
(K. Yamasaki, Y. Oki, N. Osada, M.Shimo,S. Yokoyama, S. Tokonami, A. Sorimachi, C. Kranrod and S. Takahashi)

Main Results

ARS-1: The uncertainty of external dose measurements by energy and angular response with dosimeter and worker was discussed using a passive neutron monitor consisted of CR-39 and one millimeter thickness poly-ethylene sheet. If the upper limit of confidence interval was set to 95 % of Cumulative Distribution Function (CDF), in the case of KURRI linac neutron field, the dosimeter value should be set the value of 30 to 50 percent to conform the dose limit of 50 mSv/y.

ARS-2:The concentration of radioactive particles containing of exhaust gases from accelerator for BNCT was continuously measured using a radioactive gas monitor. The concentration of exhaust gas was higher continuously for the limited period of the working of BNCT. The concentration of atmosphere was not affected by the working of BNCT. The tritium compounds from radioactive particles containing of exhaust gases from BNCT appear to be not detected and is considered to be Ar-41.

ARS-3: In the electron LINAC experiment, the soluble (0-3 nm) and colloidal (3-7, 7-16, 16-200,200- nm) concentrations of Cu and Fe were measured after irradiation time for 2 h and 12h. Dependence of photon intensity on the size profiles showed that high-energy photons promote the transfer of Cu, Fe into water phase. At the ^{60}Co gamma-ray irradiation to Cu powder/water samples, dominant Cu species in water were colloidal species, which were identified to the copper oxides (Cu_2O, CuO) by XRD analysis.

ARS-4: The screen-type diffusion battery consists of a stack of stainless steel wire screens and a backup filter. The particle size distribution is calculated from the activity of the backup filter. The size is overestimated when the radioactive gas is adsorbed on the backup filter. The influence of the adsorption was very high. The particle size is overestimated to be two times or more the correct size in the case of aerosols with 50-nm geometric mean diameter.

ARS-5: A comparative measurement of the size distributions were conducted in Linac and Cyclotron. Main artificial radio-nuclide was N-13 in the Linac target room. Owing to its relatively long time of operation, observed geometrical mean diameters (GMD) were relatively large and varied with the time of operation. On the other hand, owing to very short operation time of Cyclotron, observed radio-nuclides Mg-27 and Na-24 had very small GMD. It is clarified that GMDs based on the volumes are proportional to operation period of accelerator.

採択課題番号 23P5　　加速器施設における放射線安全管理に関する研究　　　プロジェクト
（京大・原子炉）　高橋千太郎

PR5-1 Development of Passive Neutron Dosimeter With Good Energy Response

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Radiation workers exposing to neutrons should be controlled by the radiation safety regulation. Personal dose of radiation workers should be monitored and adequately estimated. When energy of neutron distributes over ten-digit range, in addition when incident angle to workers is not stable, then existing personal dosimeters could not succeed in realistic dosimetry. This study aims to discuss a new radiation control method using a CR-39 personal dosimeter in neutron fields with wide energy range. Basic experiments on a neutron personal passive dosimeter using CR-39 combined with polyethylene had been studied. Multiple radiators structure has been designed in order to elevate its detection sensitivity to neutrons with wide energy range. Detection efficiency of CR-39 to neutrons depending on incidence angles has been simulated and the calculated results fit our experiment data. In addition, dosimetric method has been developed for unidentified situations on incident angle of neutron to personal detectors. Combination of these items succeeds in calculation of probabilistic density distribution of whole body dose for each energy range of neutron. The result is multiplied by real energy spectrum under an exposure situation. This leads to probabilistic distribution of whole body dose in each neutron work place.

Figure 1 indicates a probabilistic density distribution of whole body dose of each energy range of neutron when indication dose value of a personal dosimeter is 30 mSv, for example. Probabilistic distribution of whole body dose in a target radiation field can be calculated after multiplying the probabilistic density distribution with real neutron spectrum information. Figure 2 indicates an accumulated distribution function of whole body dose for neutrons. This figure shows that the probability of a real effective dose being less than 50 mSv is about 30 % when a personal dosimeter indicates 30 mSv. We conclude that dose limit of radiation workers can be fully kept when the alarm level or reporting level of the personal dosimeter is set as the value of 30 to 50 % of effective dose limit value determined by the radiation safety regulation. This certainty probability is estimated as about 90 to 95 %. The method we have developed realizes and ensures more adequate personal dose control of radiation workers using neutrons.

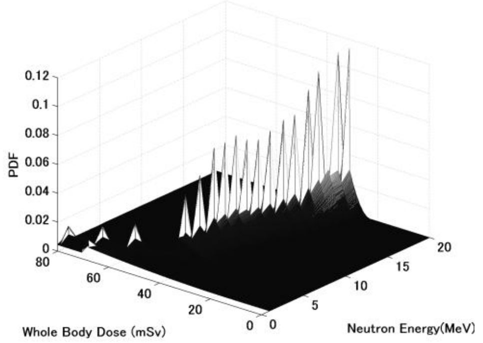

Fig. 1 . A probabilistic density distribution of whole body dose of each energy range of neutron when indication dose value of a personal dosimeter is 30 mSv.

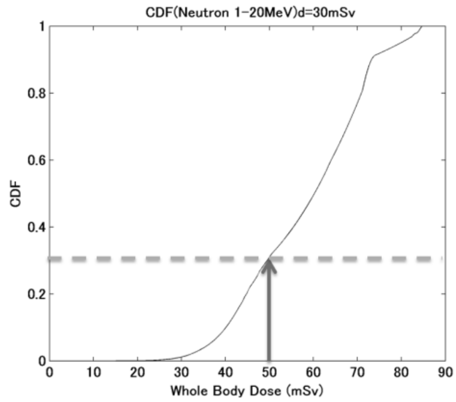

Fig.2. An accumulated distribution function of whole body dose for neutrons under the indication of 30mSv.

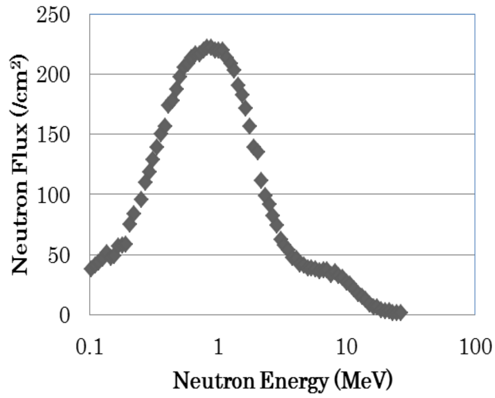

Fig.3. Neutron Spectrum from Ta target by the KURRI-LINAC.

採択課題番号 23Ｐ5-1　エネルギーレスポンスの良好なパッシブ型中性子線量計の開発　プロジェクト
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（京大・原子炉）山崎敬三、高橋知之

PR5-2

Production and Behavior of Tritium in Accelerator Laboratory

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INTRODUCTION

Radioactive particles are known to generate in the working field of accelerator. Then, the concentration of radioactive particles in the working field of Fixed Field Alternating Gradient Accelerator (FFAG) is necessary to be measured before and after working FFAG. However, the concentration of gaseous radioactive particles exhausted from working FFAG in 2011 was too low to be measured as monitoring gas. The present work was measured the concentration of radioactive particles containing of exhaust gases from the accelerator for boron neutron capture therapy (BNCT).

EXPERIMENTAL

Gaseous radioactive particles were provided from the exhaust gas from the accelerator for BNCT. They were filtered with pre filter and heap filter, and then were used as monitoring gas. The concentration of exhaust gases from BNCT, the gas permeated into Vycor glass wall used as gas separator and the atmosphere outside Innovation Research Laboratory was used as reference gas were measured alternately.

A activity of gaseous radioactive particles was measured by using radioactive gas monitor (Ohkura Electric Co., RD-1,200) and ion chamber (Ohkura Electric Co. I-409602, 1,000cm^3). The output of ion chamber was digitally recorded by a detalogger (Graphtec Co., midi LOGGER GL200).

RESULTS

The concentration of radioactive particles containing of exhaust gases from Accelerator for boron neutron capture therapy (BNCT) was continuously measured from October 2 (Sunday) in 2011 to October 8 (Saturday) in 2011.

Figure 1 shows the radioactive particles concentration of exhaust gases from BNCT, the gas permeated into Vycor glass wall used as gas separator and the atmosphere outside Innovation Research Laboratory on October 22 (Tuesday) in 2011. The radioactive particles concentration of exhaust gas was higher continuously for the limited period of the working of BNCT. The alternate radioactive particles concentration of the gas permeated into Vycor glass wall used as gas separator and the atmosphere outside Innovation Research Laboratory increased slightly due to the working of BNCT.

The permeability of a trace of tritium or argon in nitrogen into Vycor glass wall was measured. Tritium permeated fast but argon did slowly into Vycor glass wall. Therefore, The radioactive particles containing of exhaust gas is considered to argon-41.

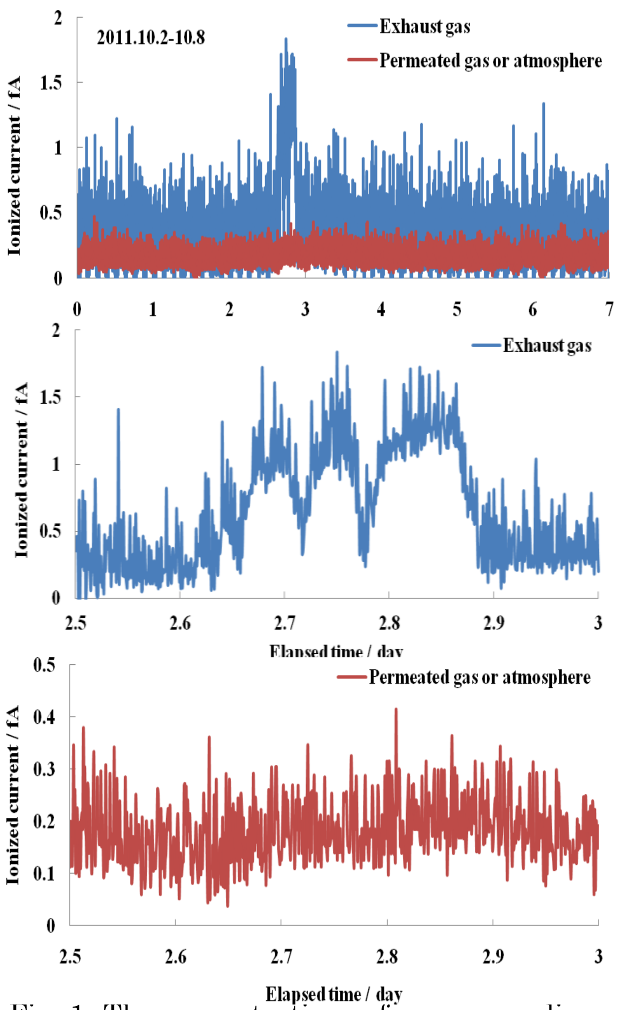

Fig. 1. The concentrations of gaseous radioactive particles contained in the exhaust gas of BNCT and atmosphere outside Innovation Research Laboratory.

採択課題番号 23P5-2　　　　加速器施設内外のトリチウム生成・動態 3　　　　プロジェクト
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PR5-3 Corrosion of Metals and Colloid Formation in Water under Intense Raiation Field

K. Bessho, H. Matsumura, K. Masumoto, Y. Oki[1], N. Akimune[2], S. Sekimoto[1], K. Yamasaki[1] and N. Osada[2]

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INTRODUCTION: At high-intensity accelerator facilities, it is possible that intense radiation fields affect chemical states of elements around beam-lines and targets. In the cooling-waters for magnets and targets, formation of metal-related colloid in water is one of the important subjects in the radiation managements. In this work, metals contacted with water were irradiated by bremsstrahlung and neutrons, or γ-rays, as a model environment inside accelerator facilities. Radiation effects on corrosion and colloid formation were investigated.

EXPERIMENTS:

(1) Irradiations at the electron LINAC

Irradiation was carried out using bremsstrahlung and neutrons generated by 30 MeV electron beam hitting on a Ta target (Φ50x62 mm) using an electron linear accelerator (LINAC) at the KURRI. A pair of Cu vessels (I. D.19x75mm) filled with pure water was fixed at the downstream position (0 deg: 40 mm from Ta target) and perpendicular position (90 deg: 40 mm from Ta target), as shown in Fig. 1. Electron beam currents and the irradiation times were 10 - 110 μA and 2 - 24 h, respectively. Intensity of high energy photons, fast neutrons, and thermal neutrons was monitored by activation method using $^{197}Au(\gamma,n)^{196}Au$, $^{27}Al(n,\alpha)^{24}Na$ and $^{197}Au(n,\gamma)^{198}Au$ reactions, respectively.

(2) Irradiations at the ^{60}Co γ-ray irradiation facility

Cu vessels filled with water, same size as used in the LINAC experiments, were irradiated by γ-rays using the ^{60}Co γ-ray irradiation facility, KURRI. The dose rates were 2.9 kGy/h or 22 kGy/h and irradiation times were 2 - 72 h.

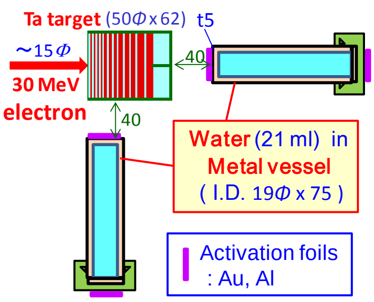

Fig. 1. Experimental setup for the irradiation of the samples at the electron LINAC.

(3) Analysis of soluble and colloidal concentrations of Cu in water after the irradiations

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

RESULTS: Figure 2 shows soluble (0-3 nm) and size-separated colloidal concentrations (3-7, 7-16, 16-200, 200- nm) of Cu in water without irradiation and after the irradiation at γ-ray irradiation facility or electron LINAC facility. The plots for the LINAC experiments in the figure are selected as the results obtained at the downstream (0-deg) position in order to evaluate the effect of photon intensity, where flux of neutrons can be neglected compared to flux of bremsstrahlung. The results in Fig. 2 demonstrate that irradiation of high-energy photons (γ-ray or bremsstrahlung) clearly affects the elution of Cu into water and formation of Cu-related colloid / particle in water phase.

Concentration of soluble Cu species increased with photon intensity. Formation of colloidal species was also noticeable at intense photon-fields. It implies that high-energy photons induce corrosion of Cu materials in water, transfer of Cu species into water phase as a soluble species or colloidal species, and growth of colloidal species in water.

Time dependence of size-profiles for Cu species formed at constant dose rates imply that dissolution of Cu metal and formation of Cu-related colloid promoted by radiation progress in the time scale of several hours to hundreds hours.

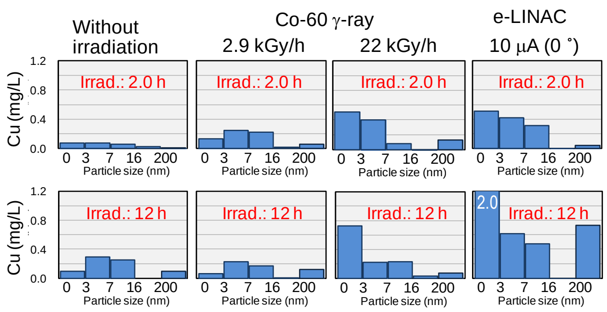

Fig. 2. Soluble (0-3 nm) and colloidal concentrations (3-7, 7-16, 16-200 nm) of Cu in water without irradiation, after the irradiation at Co-60 γ-ray irradiation facility, and after the irradiation at electron LINAC. Irradiation time : 2.0 h, 12 h

採択課題番号 23P5-3　　加速器冷却水中における放射性核種およびコロイド　　プロジェクト
化学種の挙動解析３

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（京大院・工）秋宗尚弥、長田直之

PR5-4 Adsorption of Radioactive Gas on a Membrane Filter in an Electron Linear Accelerator

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INTRODUCTION: Membrane filters are widely employed for air filtration. They are suitable for collection of radioactive aerosols and the following radioactivity measurement. However, some material of the filters collects gaseous compounds by adsorption in addition to the particulate matter. The adsorption interferes correct estimation of radioactivity of radioactive aerosols.

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. They are contained in both gaseous molecules and aerosol particles. Both species are formed through radiolysis processes of the air. Especially in electron linear accelerators (linacs), radioactive gaseous nitrogen species, such as nitric oxides and nitric acid, are dominantly formed in the air. Therefore, corrosion of accelerator hardware takes place in the accelerator rooms. Besides the corrosion problem, the information on radioactive airborne species is also important for protection of internal exposure of workers and design of exhaust systems of the facilities. The membrane filters are used in the size measurement of the aerosols, such as diffusion battery or cascade impactor. The particle size is indispensable information to estimate correct internal doses.

In this work, collection efficiency of the gaseous species of ^{13}N was estimated to correctly measure the activity of the aerosols using the radioactive gas formed in an electron linac.

EXPERIMENTS: The irradiation experiment was carried out in the 46-MeV electron linear accelerator of Research Reactor Institute, Kyoto University. In this work the adsorption of radioactive gas was estimated for Millipore HA filter (a mixture of cellulose acetate and cellulose nitrate, pore size 0.45 μm). A tantalum target was bombarded with a 30-MeV electron beam in the irradiation room. The beam current was ca. 100 μA. During the irradiation, the air was exposed to bremsstrahlung of maximum energy of 30 MeV and neutrons which were produced in the target.

The Irradiated air was sampled from the irradiation room, and was introduced to a membrane filter assembly at the flow rate of 20 L/min. The filter assembly was comprised of a PTFE (polytetrafluoroethylene) filter, without adsorption ability of radioactive gas, and the Millipore filter. Their diameter was 47 mm. After the particulate fraction was removed by the PTFE filter (pore size, 0.8 μm), the gas fraction was passed through the Millipore filter.

The activity of the filters was measured 10 min after the end of the sampling. The 511-keV annihilation γ-rays from the e^+-emitters were measured with a coincidence system of two bismuth germanate (BGO) detectors. The activity of ^{13}N was determined by analyzing decay curves of the activity of the filters.

RESULTS AND DISCUSSION: The radionuclide detected on the filters was found to be ^{13}N and almost all ^{15}O was decayed at the time of 10 min after the end of sampling. Unlike high-energy proton accelerator, carbon-11 was not observed in air the 30-MeV electron irradiation. The efficiency was expressed as the activity ratio of the gas collected on the Millipore filter to the aerosol particles on the PTFE filter. The efficiency was found to be 65 %.

The screen-type diffusion battery consists of a stack of stainless steel wire screens and a backup filter. After the aerosol particles in the sample air penetrate the screens, all the particles are collected on the backup filter. The percentage of particles which can pass through the stack of the screens is governed by a function of particle size, screen mesh size, the number of screens and other parameters. The particle size distribution is, therefore, calculated from the activity of the backup filter. The size is overestimated when the radioactive gas is adsorbed on the backup filter. The influence of the adsorption was estimated as shown in Fig.1. [1] As the obtained efficiency in this work was very high, the particle size is overestimated to be two times or more the correct size in the case of aerosols with 50-nm geometric mean diameter.

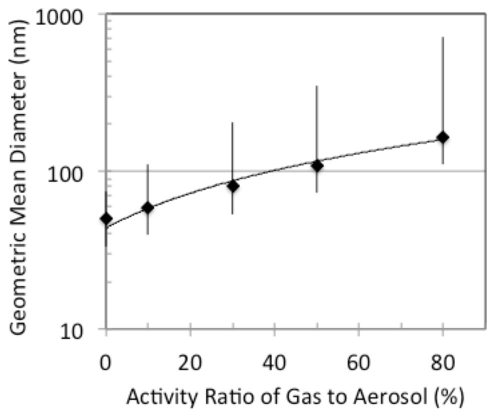

Fig. 1. Influence of the gas adsorption for 50-nm particles on the particle size measurement with the diffusion battery.

REFERENCES:

[1] N. Osada, Y. Oki, K. Yamasaki and S. Shibata, Prog. Nucl. Sci. Tech., **1** (2011) 483-486.

採択課題番号 23P5-4　　大強度加速器内で発生する放射性ガスの性状測定 3　　　プロジェクト
（京大・原子炉）沖　雄一、関本俊、山崎敬三、丸橋晃、田中浩基（京大院・工）長田直之、（藤田保健衛生大医療科学部）横山須美、（保健衛生大院保健学研究科）青木克憲

PR5-5 Varieties of Radio-Nuclides and Size Distributions of Radioactive Aerosol Produced in Different Types of Accerelators III

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Research Reactor Institute, Kyoto University
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[3]National Institute of Radiological Sciences

INTRODUCTION: The radiation protection toward the induced airborne particulate radio-nuclides is a key issues for radiation safety of the workers and maintenance of the accelerator facilities. Ultra fine particle generation and particulate radioactivity in high energy and high dose accelerator have been examined using electron Linac in this institute [1]. Recently, a proton cyclotron for BNCT has been installed. This report describes mainly the experimental results which were obtained at both accelerators.

EXPERIMENTS: Exposure experiments were carried out in the ambient air near target (Ta:Linac, Be:Cyclotron). Both accelerators were operated at the conditions of maximum energy 30 MeV and maximum mean current 90 μA to 1mA. Sampling points were about 1 m apart from the target in both cases. At the linac, sample air was introduced to the experimental room with flexible tube made of stainless steel of 2.5 cm dia. and about 6 m in length through radiation shield of 2.5 m thick concrete. At the cyclotron, sample air was introduced to the basement room using flexible tube made of charge proofing rubber of 1.2 cm dia. and about 20 m in length through a labyrinth. Sampling flow rate was about 25 ℓ /min. Number and activity-weighted size distributions which were induced high dose and high energy accelerator radiation were measured using SMPS (TSI, Model 3936), CPC (TSI, Model 3025A), laser particle counter (RION, KC-18) and low pressure cascade impactor (Tokyo Dylec, LP-20RS)

The aerosol attached N-13, Mg-27, Na-24 and Rn-progeny were sampled on the 13 stages of the impactor for thirty minutes with flow rate of 22.2 ℓ /min. The beta ray from N-13, Mg-27, Na-24 and alpha particles from Rn-progeny were sequentially measured with an automatic sample changer (Aloka, Model JDC-551S) equipped with ZnS(Ag) scintillation counter and GM counter for 1 minute after 15 minutes waiting for the decay of ^{218}Po. After decay correction, number fractions of radioactive particles were found for each stage of the impactor. Since the size distribution was close to a log-normal distribution, the data of the cumulative fraction vs. the aerodynamic diameter were plotted on a logarithmic probability section paper. The smooth line through those data points was used to find the geometric median diameter (GMD, d_g), and geometric standard deviation (GSD, σ_g).

RESULTS and DISCUSSION: Table 1 shows typical time variation data of the measured radio-nuclides and their size distributions in the electron Linac and the proton Cyclotron in KUR. Both data seems to be somewhat different. Here, N-13 aerosols which exist in the target room of the Linac are considered to coexist with largest amount of gaseous N_2 and N-13. On the other hand, Mg-27 and Na-24 are considered to be produced through nuclear reactions of high energy neutron and main constitutes of Al near the target material Be in the proton Cyclotron.

REFERENCE:

[1] S. Yokoyama *et al.* Jpn. J. Health Phys. 43(4), 2008, 333-340

Table 1. Typical time variation data of the measured radio-nuclides and their size distributions in the electron Linac and the proton Cyclotron in KUR.

Elapsed Time from Beam "ON"	Electron Linac		Proton Cyclotron		Remarks
	^{13}N		^{27}Mg ^{24}Na		
hrs	d_g(μm)	σ_g(－)	d_g(μm)	σ_g(－)	
0.17			0.083	1.70	Target Room
0.50	0.11	2.45			Chamber
1.3	0.21	1.46			Target Room
16.7	0.40	1.73			Target Room
38	0.48	1.77			Target Room

採択課題番号 23P5-5 高エネルギー放射線場で発生する放射性ナノ粒子の測定・評価 3　プロジェクト
（京大・原子炉）山崎敬三、沖雄一（京大院・工）長田直之（藤田保健大）下　道国、横山須美
（放医研）床次真司、反町篤行、C. Kranrod（京大・原子炉）高橋千太郎