I-1. PROJECT RESEARCHES

Project 9

PR9

Production Mechanism of Radioactive Aerosols Released from Fukushima Daiichi Nuclear Power Plant

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OBJECTIVE: The high amount of radioactive materials was released in the severe accident of Fukushima Daiichi Nuclear Power Plant. Investigating the chemical and physical properties of the radioactive materials in the air and soil samples are one of the most important problem to understand the exposure dose at the time of the accident and in the future and have been studied by various method. However, the production mechanism of radioactive aerosols generated in the reactor buildings does not be clarified because a part of the inside of the reactor building was under the condition of high temperature, high dose rate, etc. But elucidating the production mechanism of radioactive aerosols inside of the reactor buildings is important to understand the chemical and physical properties of radioactive materials just after the release from the reactor buildings. In addition, understanding the production mechanism will play an important role in decommissioning of the reactors because the environment in the reactor building is under quite unusual condition as used to be. In this project research, the production mechanism and measurement technique of such radioactive aerosols have been investigated experimentally.

RESEARCH SUBJECTS: The project research is composed by three individual subjects in this year period. One is developments of production method of radioactive aerosols for simulation experiments to investigate the production mechanism of radioactive solution aerosols, one is development of measurement technique of radioactive aerosols under high dose condition, and another is investigation of insoluble radioactive micro particles. The respective subjects of the research groups of this project are described as follows;

P9-1: Development of production method of radioactive aerosols by attaching radioisotopes to aerosol particles
P9-2: Development of measurement method of radioactive aerosols under severity conditions
P9-3: Investigation for production process of insoluble radioactive particles released from FDNPP

RESULTS: In the subject P9-1, experimental method using aerosol generating apparatus combined with neutron irradiated UO_2 has been developed to observe the attachment behavior of fission products of thermal velocities to solution aerosol particles. Neutron irradiated UO_2 powder was used as a source of fission products. The fission products were mixed in a cylindrical chamber with solution aerosol generated by an atomizer filled with 0.01 M sodium chloride solution. In the chamber, fission products attached to the aerosol particles to generate radioactive solution aerosol particles. The radioactive aerosol particles were collected on a filter. The gamma-ray spectrometry for the filter was performed to estimate the attachment ratio of fission products to the aerosol particles. It was found that there exist various attachment ratios depending on chemical properties of fission products. The results obtained in the present work is correspond with the previous work summarizing that attachment ratio might depend on electro static interaction between fission products and localized anions at the surface of solution aerosol particles.

Continuing from the previous year, a newly assembled screen-type diffusion battery (SDB) system was applied to measure aerosol particle size in the target room of an electron linear accelerator (LINAC) facility for the purpose of development of measurement method of radioactive aerosols under severity conditions (P9-2). An irradiation chamber was placed at a rear position of a platinum target of LINAC. The size distribution of the radiation-induced aerosols generated in the chamber was also measured by using a scanning mobility particle sizer (SMPS) in advance. The irradiated air containing the aerosol was simultaneously introduced to the SDB cylinder and the compensation line. The irradiated air was sampled continuously from both the upstream and downstream positions of the SDB cylinder, and the penetration ratios were estimated. The number concentrations of the aerosols were mainly measured with a condensation particle counter. The penetration ratios obtained by changing air flow rate were fitted to a theoretical function for lognormal distributions to obtain the geometric mean and geometric standard deviation of particle diameter. In order to confirm the performance of newly developed system, the SMPS was used for comparison with the SDB system. As a result, very good agreement is found in the particle size distribution obtained by both methods, and the performance is confirmed.

In the subject P9-3, morphological observation and elemental analysis for a radioactive insoluble micro particle which was collected from a soil sample around the power plant were performed. The particle was extracted from a soil sample by using an imaging plate at University of Tsukuba. Morphological observation was performed by SEM, and the EDX measurement was performed for particle surface to analyze the elemental composition of the particle. The whole shape is distorted, and the length of the longer direction is about 500 μm. The surface is almost smooth but has small holes partly. Concentrations of main elements are determined by EDX measurement to be 66.4, 18.8, 9.4, 1.8, 1.7 and 1.4% for oxygen, silicon, sodium, calcium, aluminum and magnesium, respectively. The main composition of the particle is thought to be silica dioxide from the elemental analysis, and element ratio is similar to that of basalt concrete containing sodium. In the near future, the generation process was experimentally simulated on the basis of obtained chemical composition to understand the generation process of insoluble radioactive micro particles.

PR9-1 Electrostatic Interaction in Production Process of Radioactive Solution Aerosol Particles

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INTRODUCTION: Elucidating generating process of radioactive aerosols is one of the most important problem to understand the behavior of radioactive aerosols released from the Fukushima Daiichi Nuclear Power Plant after the Great East Japan Earthquake in 2011. The generating process has been investigated using an aerosol generating apparatus combined with a spontaneous fission source of ^{252}Cf in our previous work [1-4]. It was found that there are two types of attachment processes of fission products to aerosol particles; one is caused by geometric collision and another is induced by electrostatic interaction between a fission product and surface of an aerosol particle. And dependence of species and concentration of solute on attachment behavior of fission products to solution aerosol particles has been found in the previous study. However, the electrostatic interaction could not be quantitatively clarified because fission products emitted from ^{252}Cf source have recoil velocities which cause the attachment process by geometric collision to increase. Experiments similar to the previous study using fission products with thermal velocity might elucidate the electrostatic interaction qualitatively. In the present work, experimental method using aerosol generating apparatus combined with neutron irradiated UO_2 has been developed to observe the attachment behavior of fission products with thermal velocity to solution aerosol particles.

EXPERIMENTS: The neutron irradiation to uranium dioxide was carried out at Kyoto University Research Reactor (KUR). Powder of UO_2 was encapsulated in a quartz tube under reduced pressure, and the quartz tube covered by a polyethylene tube was inserted into polyethylene capsule to irradiate neutrons using pneumatic transport system (Pn-2) of KUR. The amount of UO_2 was 10 mg, and the neutron irradiation time is 30 min. The experimental setup for production of radioactive aerosol is shown in Figure 1. A part of irradiated UO_2 powder was extracted to another quartz tube placed in an electric furnace. Fission products produced in the irradiated UO_2 powder was released by heating the furnace up to 1100°C. On the other hand, Atomizer filled with 0.01 M sodium chloride solution generate solution aerosol. Both released fission products and solution aerosol particles were aspirated by a suction pomp to be transported and mixed in a cylindrical chamber. And radioactive aerosol particles were produced by attaching fission products to aerosol particles in the chamber. The produced radioactive aerosol particles were collected on a polycarbonate filter at downstream of the chamber. The amount of fission products which attaches to aerosol particles were estimated by gamma-ray spectrometry for the filter using a Ge-detector. On the other hand, the amount of fission products released from the irradiated UO_2 powder by heating was estimated by subtraction of gamma-ray spectra for before and after heating the UO_2 powder.

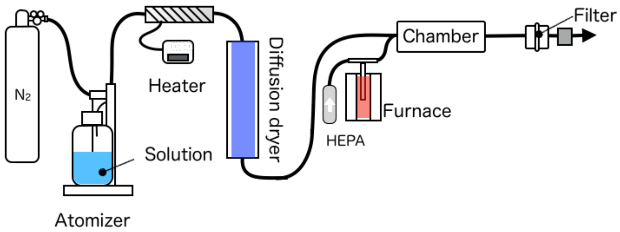

Fig. 1 Experimental setup for generating radioactive solution aerosol using neutron irradiated UO_2.

RESULTS: The attachment ratio of fission products to aerosol particles was estimated as shown in Figure 2. It was found that there exist various attachment ratios among various fission products. Fission products which can forms positively-charged chemical species (Zr, Te and Ce) might show higher attachment ratio compared with those form negatively-charged or neutral species (Ru, I and Xe). These results correspond with our previous work summarizing that attachment ratio might depend on electro static interaction between fission products and localized anions [5] at the surface of solution aerosol particles.

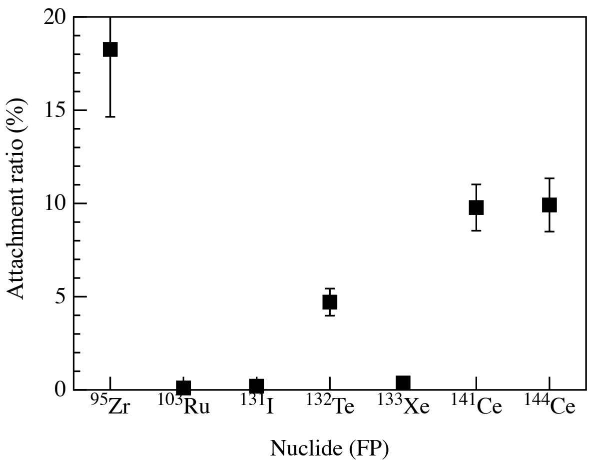

Fig. 2 Attachment ratio of FP released from neutron irradiated UO_2 to 0.01 M NaCl solution aerosol particles.

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PR9-2 Particle Size Measurement of Radioactive Aerosol Particles in An Electron LINAC Using A Diffusion Battery System II

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INTRODUCTION: Radioactive nuclides are evaporated and released to the environment in target melting accidents in accelerator facilities like the J-PARC accident in 2013. The radionuclides are often incorporated into metallic aerosols emitted from the target metal or ambient aerosols in the target room. Therefore, properties of the ambient aerosols like their particle size and concentration are very important information to clarify the behavior of the radioactive nuclides.

During machine operation the target room is filled with radiation-induced aerosol particles in the size range of several nm to ca. 100 nm. The size for the radioactive particles was often measured using a wire screen technique in accelerator facilities. Convenient size measurement techniques are needed from the viewpoint of radiation protection in accelerator facilities.

In this work, continuing from the previous year [1], a newly assembled screen-type diffusion battery (SDB) system was applied to measure aerosol particle size in the target room of an electron linear accelerator (LINAC) facility.

EXPERIMENTS:
The measurement system: The SDB system used in this work consists of an SDB line (Line A in Fig. 1) and a compensation line (Line B). Each line was connected to a ball valve and a mass flow controller (MFC) to change a flow rate independently. All valves and MFCs were controlled by a PC for automatic measurement of aerosol size distribution. In the SDB line, an air-tight wire screen cylinder containing a stack of stainless steel screens (500 mesh).

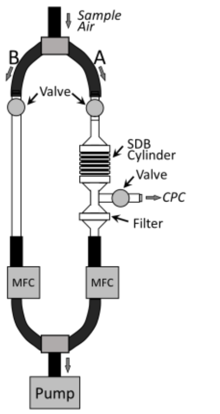

Fig. 1 The SDB system employed in this work

When very fine aerosol particles pass through wire screens, a part of the particles are trapped on the wire surface of the screens by their diffusion according to their particle size. The loss by the screens is expressed as a function of particle size, coarseness and number of screens, and flow rate of particles. The size distribution of the aerosol particles can be calculated by measuring the penetration ratio (N/N_0), where N_0 and N are number concentrations of the aerosol particles before and after penetrating screens, respectively.

Irradiation: The air-irradiation experiment was carried out in the 46-MeV electron LINAC of the Institute for Radiation and Nuclear Science, Kyoto University (KURNS). An irradiation chamber was placed at a rear position of a platinum target. During the irradiation, aerosol-free air was introduced to the chamber from the experiment room next to the target room. The target was bombarded with a 35-MeV electron beam to produce bremsstrahlung. The bremsstrahlung ionizes air and produces the radiation-induced aerosol. The beam current was ca. 40 μA.

Size measurement based on number concentration for the radiation-induced aerosol: The size distribution of the radiation-induced aerosols was confirmed to have a lognormal shape using a scanning mobility particle sizer (SMPS) in advance. The irradiated air containing the aerosol was simultaneously introduced to the SDB cylinder and the compensation line. The air flow rate for the SDB was gradually changed from 0 to 15 L/min, while that for the compensation line was decreased from 15 to 0 L/min so that the total flow rate was maintained constant not to change irradiation rate of air in the irradiation chamber. The irradiated air was sampled continuously from both the upstream and downstream positions of the SDB cylinder, and the penetration ratios were calculated. The number concentrations of the aerosols were mainly measured with a condensation particle counter (CPC).

RESULTS: The penetration ratios (N/N_0) obtained by changing air flow rate were fitted to a theoretical function [2] for lognormal distributions to obtain the geometric mean and geometric standard deviation of particle diameter. An SMPS with nano-differential mobility analyzer was used for comparison with the SDB system.

Figure 2 shows the particle size distribution obtained using the SDB cylinder with a stack of three pieces of 500-mesh screen. It was in very good agreement with that obtained with SMPS.

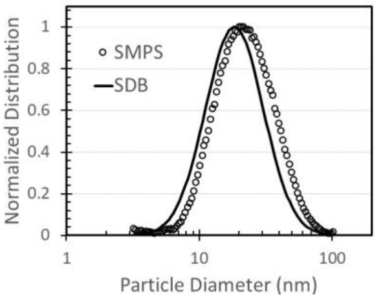

Fig. 2 Comparison of particle size distributions obtained with the SDB system and SMPS

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PR9-3 Observation of Insoluble Radioactive Microparticle Released from FDNPP

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INTRODUCTION: The characteristics of radioactive micro particles which were released from Fukushima Daiichi Nuclear Power Plant have been analyzed using various method [1-5]. The effect of the micro particles to the environment is important because the particles are mostly insoluble and a part of them has high radioactivity. And these high radioactive microparticles might exist in the reactor building even now and will affect exposed dose in decommissioning process of the reactors. Morphological observation and elemental analysis for the radioactive insoluble micro particle which was collected from a soil sample around the power plant were carried out in the present work.

EXPERIMENTS: The particle observed in the present work was extracted from a soil sample by using an imaging plate at University of Tsukuba. Morphological observation was performed by SEM (Phenom ProX, Thermo Fisher Scientific K.K.). And the EDX measurement was performed for particle surface to analyze the elemental composition.

RESULTS: The SEM image of overview of the micro particle is shown in Figure 1. The whole shape is distorted, and the length of the longer direction is about 500 µm. The surface is almost smooth but has small holes partly. The EDX measurement was performed for a part of smooth surface area to analyze the elemental composition. The obtained EDX spectrum is shown in Figure 2. Concentrations of main elements are determined to be 66.4, 18.8, 9.4, 1.8, 1.7 and 1.4% for oxygen, silicon, sodium, calcium, aluminum and magnesium, respectively. From the elemental analysis for other parts of the particle surface, similar results were obtained. The main composition of the particle is thought to be silica dioxide from the elemental analysis, and element ratio is similar to that of basalt concrete containing sodium. It is impossible to identify a generating process of the particle from the present results, however, following process is conceivable. Basalt concrete was used as pedestals supporting a reactor pressure vessel. The pedestal with sea water which was injected as coolant from outside were heated by contact with melted core, and the microparticles were generated through decomposition, melting, scattering, recooling and other process. In the near future, the generation process was experimentally simulated by heating mixture of concrete and sodium chloride to understand the generation process of radioactive micro particles.

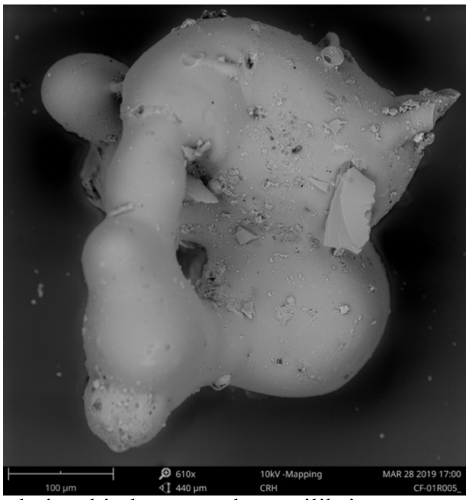

Fig. 1 Relationship between the equilibrium constant and concentration of solute.

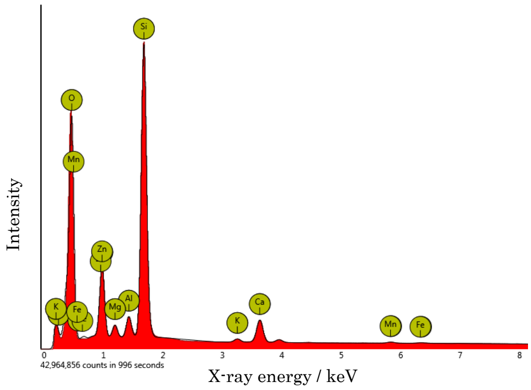

Fig. 2 EDX spectrum of a part of smooth surface of the micro particle.

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