PR8 Production Mechanism of Radioactive Aerosols Released from Fukushima Daiichi Nuclear Power Plant

K. Takamiya

Research Reactor Institute, Kyoto University

OBJECTIVE: The chemical and physical characteristics of radioactive materials released from Fukushima Daiichi Nuclear Power Plant to the atmosphere has been investigated by various methods to elucidate the environmental dynamics of the radioactive materials. A radioactive aerosol is one of the most important released materials, and the production mechanism and measurement technique has been studied in this project. The generation method for solution radioactive aerosols had been developed in the previous studies in this project, and the new subject in which the generated solution aerosol had been applied was added in this year period in order to understand the environmental dynamics of the radioactive aerosols.

RESEARCH SUBJECTS: The project research is composed by five individual subjects in this year period. Two are developments of production method of radioactive aerosols for simulation experiments, one is development of measurement method of radioactive aerosols, and one is elucidation of the production mechanism of aerosol under the specific situation. And one subject about interaction of radioactive cesium in the environment was added in this year period. The respective subjects of the research groups of this project are described as follows;

P8-1: Development of production method of radioactive aerosols by attaching radioisotopes to aerosol particles
P8-2: Development of measurement method of radioactive aerosols under severity conditions
P8-3: Development of production method of radioactive aerosols by heating radioisotopes
P8-4: Study for production mechanism of radioactive aerosols in the presence of sea water
P8-5: Interaction of radioactive cesium and suspended particles in river water

In this year period, experiments of subjects P8-3 and P8-4 were not carried out because of no operation of KUR.

RESULTS: In the subject P8-1, the solution concentration dependence of production behavior for radioactive solution aerosols has been investigated. Primary aerosols were generated from sodium chloride solution of various concentration (0.005, 0.01 and 0.02 M). The production ratio of radioactive aerosols was estimated as attachment ratio of fission products to aerosol particles in a chamber in which ^{252}Cf spontaneous fission source was placed. The attachment ratio of ^{104}Tc can be obtained only with enough statistics as a function of total surface area of aerosol particles in this experiment. It was found that there are differences among aerosols of three different concentrations in the relationship. The result might suggest the solute concentration of solution aerosols affects the production process of radioactive aerosols. The relationships were fitted by an equilibrium function on the basis of an assumption of the adsorption-desorption equilibrium between fission products and surface of aerosol particles. The order of magnitude of the estimated equilibrium constants is 0.02 M > 0.01 M > 0.005 M. This trend might be caused by density of the anions in the solution aerosol particles.

For the development of measurement method of radioactive aerosols under severity conditions (P8-2), time variation of particle size of the radiation-induced aerosols was tried to be measured in an electron linac facility by using a scanning mobility particle sizer (SMPS) or a wire screen method. According to the SMPS measurement, no aerosol particles were observed in the chamber immediately after the filtration of the irradiated air; however, rapid formation of the aerosol occurred in the chamber. It was found that the particle size increased to approximately 100 nm (geometric mean) with a decrease in the number concentration, and time to maximum size from the end of air sampling was 9 h. The size and concentration are expressed as functions of the rates of formation, coagulation, evaporation, dilution and disappearance due to collision to the inside wall of the chamber.

For the research about interaction of radioactive cesium in the environment（P8-5）, the effect of cesium adsorption to suspended particles on the desorption behavior was investigated. The suspended particles were sampled from Abukuma river in Date, Fukushima. For the adsorption of cesium, 100 mg of the particle sample was exposed by the cesium solution aerosol generated by an atomizer containing 0.4 wt% of CsCl, for 2.5 hours or 90 seconds. The amount of loaded cesium on the particles was determined by ICP-MS using a part of particle samples. And the particle samples exposed by the aerosol were mixed with various desorption agents as the cesium desorption treatment. The supernatant was filtered by a 0.45 μm pore size membrane filter, and the cesium concentration in it was measured by ICP-MS to estimate the desorption ratio. The results of desorption experiment indicated that all desorption agents (470 mM NaCl, seawater, 1 M KCl) except ultrapure water desorbed cesium more than 90 % from the high cesium loading sample. In contrast, only 30 % cesium was desorbed from the low cesium loading sample by seawater. As a result, the cesium desorption percentage by seawater was 30 %, where the cesium loading on the suspended particles was about 1μg/g which was close to the loading amount of 90 seconds aerosol atomized sample (= 6.9μg/g). The observed agreement of the desorption percentages from different way of cesium loading indicates that the desorption behavior is simply governed by the cesium loading amount on the particles rather than the way of loading.

In the near future, experiments with a neutron irradiation using KUR will be performed, and further progress is expected in this project.

PR8-1 Solution Concentration Dependence of Production Ratio of Radioactive NaCl Solution Aerosols

K. Takamiya, Y. Nishizawa, S. Sekimoto, Y. Oki and T. Ohtsuki

Research Reactor Institute, Kyoto University

INTRODUCTION: Large amount of radioactive materials released into the atmosphere from Fukushima Daiichi Nuclear Power Plant have been transported by aerosols. It was suggested that the potential transport medium for radioactive cesium were sulfate aerosols [1, 2]. On the other hand, the initial production process of radioactive aerosols in the reactor building is not clear because there is not direct measurement data. Attachment behavior of fission products to solution aerosol particles have bee studied in our previous works [3, 4] to elucidate the production mechanism of radioactive solution aerosol in the initial production process. It was found that there seems to be an adsorption equilibrium process in the radioactive aerosol production and the equilibrium constants depends on a kind of solution. In the present work, the solution concentration dependence of production behavior for radioactive solution aerosols has been investigated.

EXPERIMENTS: The production method of radioactive aerosols using a spontaneous fission source of ^{252}Cf was explained in the previous reports [3, 4]. Primary aerosols were generated from sodium chloride solution of various concentration (0.005, 0.01 and 0.02 M). In order to examine the relationship between surface area of aerosol particles and the production ratio of radioactive aerosol, the primary solution aerosol was classified by size using a differential mobility analyzer in the region of diameter smaller than about 100 μm. In the larger diameter region, the surface of the aerosols was controlled by changing the aerosol number concentrations. The radioactive aerosol produced by attaching fission products to aerosol particles were collected by a polycarbonate filter. On the other hand, all fission products emitted from the 252Cf source were collected by a grease-applied cellulose filter placed directly on the source. Gamma-ray spectrometry using a Ge-detector was performed for the filters to estimate the production ratio of radioactive aerosols which were produced by attaching fission products to aerosol particles.

RESULTS: The production ratio of radioactive aerosols was estimated as attachment ratio of fission products to aerosol particles from the ratio of photopeak areas for each fission product. The attachment ratio of 104Tc can be obtained only with enough statistics and shown in Fig. 1 as a function of total surface area of aerosol particles. The symbols of circle, square and triangle show the attachment ratio for sodium chloride solution of 0.005, 0.01 and 0.02 M solution concentrations, respectively. The difference are found among aerosols of three different concentrations, and this result means the solute concentration of solution aerosols affects the production process of radioactive aerosols. The relationships were fitted by an equilibrium function on the basis of an assumption of the adsorption-desorption equilibrium between fission products and surface of aerosol particles. The results of the fitting are shown by lines in Fig. 1. The order of magnitude of the estimated equilibrium constants is 0.02 M > 0.01 M > 0.005 M. This trend might be caused by density of the anions in the solution aerosol particles.

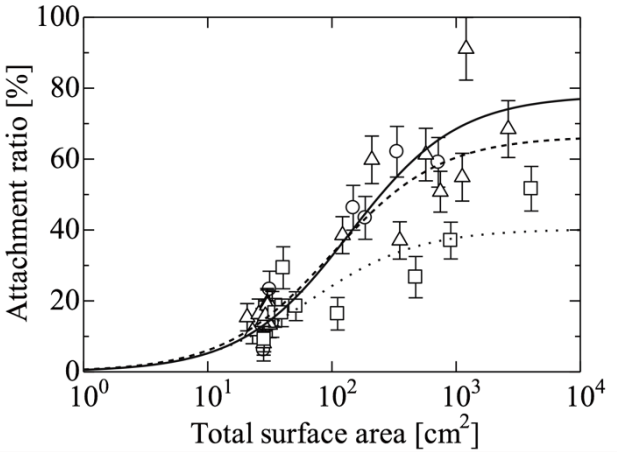

Fig. 1. Relationship between the attachment ratio of ^{104}Tc to NaCl solution aerosol and the total surface area of the aerosol particles. Circles, triangles and squares show the attachment ratio to NaCl solution aerosols of 0.005, 0.01 and 0.02 M concentrations, respectively.

REFERENCES:

[1] N. Kaneyasu et al., Environ Sci Technol **46** (2012) 5720–5726.
[2] Y. Miyamoto et al., J. Environ. Radioact. **132** (2014) 1–7.
[3] K. Takamiya, et al., J. Radiat. Prot. Res. **41** (2016) 350-353.
[4] K. Takamiya, et al., J. Radioanal. Nucl. Chem. **307** (2016) 2227-2230.

PR8-2 Growth of Aerosol Particles in Irradiated Air of an Accelerator Room

Y. Oki, K. Takamiya, N. Osada[1], S. Sekimoto and T. Ohtsuki

Research Reactor Institute, Kyoto University
[1]Advanced Science Research Center, Okayama University

INTRODUCTION: Radioactive aerosols were released to the environment in both accidents of the Fukushima Daiichi Nuclear Power Plant (FDNPP) and Japan Proton Accelerator Research Complex (J-PARC). The release occurred due to melting of the highly radioactive nuclear fuel or the metallic target. Radionuclides evaporated from the molten fuel or metals are incorporated into ambient aerosol particles to form the radioactive aerosols of various radionuclides. The radiation-induced aerosol is a principal ambient aerosol in intense radiation fields; however, most of its nature concerning formation and growth in radiation fields remain unexplained.

In this work, time variation of particle size of the radiation-induced aerosols was studied using a scanning mobility particle sizer (SMPS) from the time of their birth in an electron linac facility. In addition, a wire screen method was also applied for the size measurement [1].

EXPERIMENTS: An air irradiation experiment was carried out in the 46-MeV electron linear accelerator of the Research Reactor Institute (KURRI). A tantalum target was installed at the end of the beam line, and was bombarded with a 30-MeV electron beam to produce bremsstrahlung and neutrons. The bremsstrahlung ionizes air and produces radiation-induced aerosol. The beam current was ca. 100 μm. During the irradiation, the air ventilation in the target room was stopped not to disturb air. Air was collected from the target room to a 200-L vacuum aerosol chamber through a sampling tube penetrating a 3-m thick shielding wall. The target room air was introduced to the vacuum chamber after filtration with a HEPA filter. A 1.5-L ionization chamber was used for monitoring concentration of radioactive gases.

The particle size distribution and number concentration of the aerosols were measured continuously with SMPS. The irradiated air was sampled from the chamber to the SMPS at the flow rate of 0.3 L/min. A filtered aerosol-free air was supplied to the chamber at the same flow rate to keep pressure in the chamber.

A screen device consisting of a stack of 20 pieces of 500-mesh stainless steel wire screens was also employed for size measurement of the aerosol. The particle size distribution was calculated from penetration ratios for the screens obtained with condensation particle counter (CPC).

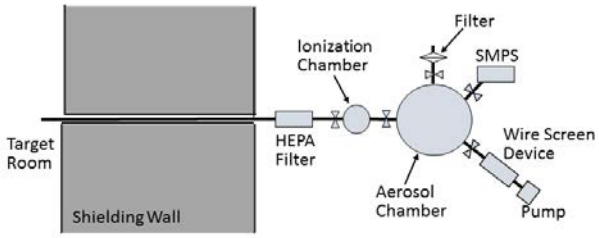

Fig. 1 Experimental Setup for Sampling of the irradiated air from the target room

RESULTS: According to the SMPS measurement, no aerosol particles were observed in the chamber imediately after the filtration of the irradiated air; however, rapid formation of the aerosol occurred in the chamber.

Preliminary results showed that the particle size incereased to approximately 100 nm (geometric mean) with a decrease in the number concentration. It took 9 h to reach the maximun size from the end of air sampling. The size and concentration are expressed as functions of the rates of formation, coaguration, evaporation, dilution and dissapearance due to collision to the inside wall of the chamber.

Oxygen-15 (half life: 2 min), ^{13}N (9.965 min) and ^{41}Ar are principal radionuclides formed in the target room air. Nitrogen-13 was a dominant gaseous nuclide in the chamber. The radioactivity in the chamber is considered to influence the formation rate. The formation rate decreased as 13N decayed.

The screen device was not applied to radioactivi-ty-based particle size measurement because conversion of radioactive ^{13}N gas to aerosols was not observed in the chamber. In the number-based particle size measurement, the number concentration of the aerosols was measured by varying the sampling flow rate to the device, not by changing number of the screens, in order to minimize the sampling volume from the chamber.

REFERENCE:

[1] Y. Oki, et al., J. Radiat. Prot. Res., 41 (2016) 216-221.

PR8-3 Interaction of Radioactive Cesium and Suspended Particles in River Water

A. Kirishima, M. Onodera, K. Takamiya[1], T. Ohtsuki[1], Y. Oki[1], S. Sekimoto[1] and N. Sato

Institute of Multidisciplinary Research for Advanced Materials, Tohoku University

[1]*Kyoto University Research Reactor Institute*

INTRODUCTION: Radioactive Cs deposited in river due to the nuclear accident of Fukushima in 2011 is thought to be adsorbed in suspended particles (SP) and migrate along the river flow. Because SP are mainly composed by clay minerals, they can retain Cs on the surface and inter-layers of clay minerals by cation-exchange reaction. When retained radioactive Cs arrives at brackish water area, they could be desorbed from SP in response to solution condition change. The understanding of this Cs speciation change mechanism is very important for the prediction of Cs migration from mountainy area to coast area. Therefore, this study carried out cesium adsorption and desorption experiment for SP by batch method. To investigate the effect of the Cs adsorption method to the particle on the desorption behavior, cesium desorption experiment from the suspended particles that adsorbed cesium by atomizing cesium aerosol was carried out in this study.

EXPERIMENTS: SP was sampled from Date place located in about 60 km northwest of the 1F NPS (Yanagawa-Ohashi, Yanagawa-cho, Date-shi, Fukushima prefecture, Japan) of Abukuma river and named as Date Sample. The river water containing suspended particles were collected about 20 L by a Teflon container. The suspended particles were then collected on a filter having 0.45 μm pore, and these particles were freeze dried. Finally, black brown color powdered suspended particles were obtained. Solution aerosols were generated using a combined system of an atomizer 3076 (TSI, Shoreview, MN) containing 0.4 wt % of CsCl, a electric furnace, and a diffusion dryer. Nitrogen gas was supplied at pressure of 2.5 kg/cm^2 to the atomizer to discharge CsCl solution aerosols [1]. The aerosols were passed through the furnace, which was kept constant at 80 ˚C and the diffusion dryer with an infill of silica gel sequentially. For the adsorption of cesium, 100 mg of the Date sample was exposed by the cesium aerosol for 2.5 hours or 90 seconds with mixing the particles every 30 minutes and 10 second, respectively by a spatula. To determine the loaded cesium amount on the suspended particles, about 4 mg of the aerosol exposed Date samples were dissolved in the mixed solution of 3 ml of conc. HNO_3 and 300 μl of 46 wt % Hydrofluoric acid in a PTFE beaker (Heatable beaker, Sanplatec Co.) with heating at 160 ℃. After the complete dissolution of the particles, the solution was dried up, then about 5 ml of 1 M HNO_3 solution was added to dissolve remained salts. The cesium concentration in this solution was then measured by an induced coupled plasma – mass spectrometry spectrometer (ICP-MS, ELEMENT II, Thermo Fisher Scientific Co.) using tellurium ion as the internal standard. These dissolution and measurement procedure were repeated 4 times for one sample of the suspended particles to get an average value of the cesium concentration. The loaded cesium concentration of the Date sample from the cesium aerosol was thus calculated from the measured cesium concentration by ICP-MS. Subsequently, the Date samples exposed by the aerosol were mixed with desorption agents in a solid-to-liquid ratio of 0.17 g/l for 6 days at 25 ℃ as the cesium desorption treatment. The supernatant was then, filtered by the 0.45 μm pore size membrane filter, and the cesium concentration in it was measured by ICP-MS. Finally the desorption percentages were calculated by following equation:

$$\text{Desorption [\%]} = (C/C_0) \times (S/L) \times 100, \qquad (1)$$

where C_0 is initial cesium concentration (g/g) in the Date sample exposed by the aerosol, and C is cesium concentration (g/l) in the supernatant solution after the desorption treatment, (S/L) is solid-to-liquid ratio (g/l) at the desorption treatment.

RESULTS: The loaded cesium amount on the Date samples atomized by the CsCl aerosol were determined to be 113 μg/g for 2.5 hours of the atomization sample and 6.9 μg/g for 90 seconds atomization by ICP-MS. The results of desorption experiment indicated that all desorption agents (470 mM NaCl, seawater, 1 M KCl) except ultrapure water desorbed cesium more than 90 % from the high cesium loading sample (2.5 hours atomization). In contrast, only 30 % cesium was desorbed from the low cesium loading sample (90 seconds atomization) by seawater. In the desorption experiment using cesium adsorbed particles from the solution, the cesium desorption percentage by seawater was 30 %, where the cesium loading on the suspended particles was about 1μg/g which was close to the loading amount of 90 seconds aerosol atomized sample (= 6.9μg/g). The observed agreement of the desorption percentages from different way of cesium loading indicates that the desorption behavior is simply governed by the cesium loading amount on the particles rather than the way of loading.

REFERENCE:

[1] K. Takamiya, T. Tanaka, S. Nitta, S. Itosu, S. Sekimoto, Y. Oki, T. Ohtsuki, Attachment Behavior of Fission Products to Solution Aerosol. J Radiat Prot Res, (2016) 41, 350–353.