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

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OBJECTIVE: Environmental distribution and transition of radioactive materials released from Fukushima Daiichi Nuclear Power Plant has been investigated by means of air dose rate measurements and radioactivity measurements for environmental samples. On the other hand, chemical forms of the radioactive materials released from the power plant and transition mechanisms in the environment has not been revealed. The chemical form of the radioactive materials just behind the release is important to evaluate the internal exposure by radioactive aerosols and the transition of the materials in soils and plants after the fall out. A radioactive aerosol is one of the most important transport medium of the radioactive materials. In the present project research, the production mechanism of radioactive aerosols has been studied to elucidate the chemical form of the radioactive materials released from the power plant.

RESEARCH SUBJECTS: The project research is composed by four individual subjects in this year. 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. The respective subjects of the research groups of this project are described as follows;
P10-1: Development of production method of radioactive aerosols by attaching radioisotopes to aerosol particles
P10-2: Development of measurement method of radioactive aerosols under severity conditions
P10-3: Development of production method of radioactive aerosols by heating radioisotopes
P10-4: Study for production mechanism of radioactive aerosols in the presence of sea water

In this year period, experiments of subjects P10-3 and P10-4 were not carried out because of no operation of KUR.

RESULTS: In the subject P10-1, an attachment ratio of fission products to aerosol particles was estimated using a solution aerosol production apparatus and a spontaneous fission source of ^{252}Cf in order to elucidate the attachment mechanism of fission products to aerosol particles.

The solution aerosols were generated from alkali halide (NaCl, NaBr, NaI, CsCl) and ammonium sulfate solution of 0.1 wt% concentration using an atomizer. The attachment ratio can be considered by associating with surface area of aerosol particles definitely because aerosols classified by size were used in the experiments. The relationships between the surface area and the estimated attachment ratio for a fission product of ^{104}Tc were fitted by a function on the basis of the assumption of the adsorption-desorption equilibrium between fission products and surface of aerosol particles. From results of the fitting, equilibrium constants and saturated attachment ratios were obtained for the combination of ^{104}Tc and each solution aerosol. In the comparison among the estimated equilibrium constants of ^{104}Tc for solution aerosols, it was found that the equilibrium constant for sodium chloride solution aerosol is smaller than those for other solution aerosols. This difference might be caused by density profiles of the anions in the fine solution particles that is explained by molecular dynamics simulations.

For the development of measurement method of radioactive aerosols under severity conditions, an air irradiation experiment was performed using the FFAG (Fixed Field Alternating Gradient) proton accelerator, and a combination method of imaging plate (IP) measurement and the wire screen technique was tried to estimate particle size of ^{11}C-bearing aerosols which were produced in the air irradiation. The ^{11}C activities collected by each screen and the backup filter were estimated from the decay analysis. The activity ratio of particles penetrating 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. Log-normal distributions are assumed as the size distribution function in the calculation because the number-based size distribution was confirmed to be log-normal by another measurement method. The geometric mean diameter for the ^{11}C-bearing aerosol particles was successfully obtained using the combination method of the wire screen collection and the IP measurement. The activity-based geometric mean diameters were larger than the number-based one by ca. 20 nm. This shows that attachment process of radioactive atoms to the surface of aerosol particles is included in the radioactive particle formation.

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Adsorption-desorption Equilibrium in Attachment of Fission Product to Solution Aerosol

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INTRODUCTION: Almost radioactive materials released in the air from Fukushima Daiichi Nuclear Power Plant have been transported by aerosols. Measurement results about size distributions and activity size distributions of the radioactive aerosols have been reported, and it was suggested that sulfate compounds were the potential transport medium for radioactive cesium isotopes [1, 2]. However, the reason for the selectivity to sulfate compounds in production mechanism of radioactive aerosols is not clear. Attachment behavior of fission products to solution aerosol particles has been investigated in order to elucidate the production mechanism of radioactive aerosols. The aerosols which contains a radioactive fission product were generated and attachment ratio of fission product aerosol was measured to reveal the production mechanism of the radioactive solution aerosols in the present work.

EXPERIMENTS: The fission-product aerosols were produced by passing a primary solution aerosol through a chamber in which ^{252}Cf source was placed. The ^{252}Cf source was covered by two Havar foils which thickness is 2.5 μm each to reduce kinetic energy of fission products emitted from the source. Primary aerosols were generated from alkali halide (NaCl, NaBr, NaI, CsCl) and ammonium sulfate solution of 0.1 wt% concentration using an atomizer. The primary solution aerosol was classified by size using a differential mobility analyzer before the injection to the ^{252}Cf chamber. The radioactive aerosol was produced in the chamber by attaching fission products to aerosol particles and collected on a polycarbonate filter. Radioactivity of fission products attaching to aerosol particles was measured by gamma-ray spectrometry using a Ge detector. Fission products emitted in the chamber from the ^{252}Cf source were also collected on a cellulose filter placed directly on the source and measured by the Ge detector.

RESULTS: The attachment ratio of fission products to aerosol particles was estimated from the ratio of photopeak areas for each fission product and shown in Fig. 1 as a function of total surface area of aerosol particles. The attachment ratio seems to be proportionate to the surface area of aerosol particles in the small surface area region. The increasing rate of the relationship varies with materials of the aerosol particles. The proportional relation and the variation of increasing rates suggest that the attachment ratio of fission products to aerosol particles

depends on both geometric collision rates and chemical effects. On the other hand, the increasing rates decrease and the attachment ratios were saturated in the larger region of the surface area of aerosol particles. This trend indicates that the attachment behavior of fission products to aerosol particles could be held in an adsorption-desorption equilibrium.

The relationships between the surface area and the attachment ratio for ^{104}Tc were fitted by a function on the basis of the assumption of the adsorption-desorption equilibrium between fission products and surface of aerosol particles. As results of the fitting which are shown by solid lines in Fig. 1, equilibrium constants and saturated attachment ratios were obtained for the combination of ^{104}Tc and each solution aerosol. In the comparison among the estimated equilibrium constants of ^{104}Tc for solution aerosols, it was found that the equilibrium constant for sodium chloride solution aerosol is smaller than those for other solution aerosols. This difference might be caused by density profiles of the anions in the fine solution particles that is explained by molecular dynamics simulations [3, 4].
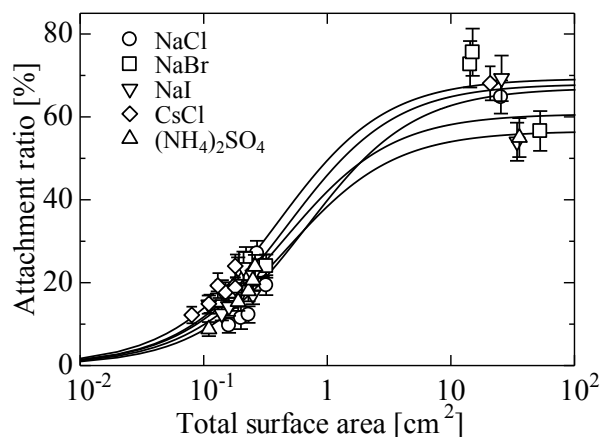

Fig. 1 Attachment ratio of ^{104}Tc to various solution aerosol as a function of total surface area of aerosol particles.

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PR10-2 An Application of Imaging Plates to Size Measurenent of Radioactive Aerosol Particles Produced in Accelerator Room Air

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INTRODUCTION: Very fine radiation-induced aerosol particles are often observed in air of accelerator rooms during machine operation. The particles are produced by an air ionization process by primary beams or by charged particles emitted from beam loss points. The particles incorporate radioactive atoms to form radioactive particles. The size for 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.

When aerosol particles pass through a stack of wire screens, a part of the particles are trapped on the screens according to their particle size. As the loss by the screens is expressed as a function of particle size, coarseness of screens, and flow rate of particles, the particle diameter can be calculated by measuring the loss by each screen.

In this work, air irradiation experiment was performed in the FFAG (Fixed Field Alternating Gradient) proton accelerator in the Research Reactor Institute (KURRI), and a combination method of imaging plate (IP) measurement and the wire screen technique was applied to estimate particle size of ^{11}C-bearing aerosols.

EXPERIMENTS: A screen device consisting of a stack of stainless steel wire screens and a PTFE backup filter was used for size measurement of the radioactive aerosol. The stacking order and number of the screens are single 100, 200, 300-mesh screens and 40 pieces of 500-mesh screens in a direction toward the downstream.

An air-irradiation chamber was installed at the upstream of the beam dump of the beam extraction line of the FFAG main ring (Fig. 1). The chamber was irradiated for 1 h with a stable beam of nominal 150-MeV protons. The beam current was 1.8 nA. During the irradiation aerosol-free air was continuously introduced at the flow rate of 8.1 L/min to the chamber, and the irradiated air was sampled with the screen device at the rate of 7.5 L/min. A part of the irradiated air was introduced to an SMPS (Scanning Mobility Particle Sizer) to monitor the number-based aerosol size simultaneously with the screen collection.

In the aerosol the radionuclides affecting the IP exposure are virtually only positron emitters such as ^{15}O (half life: 2 min), ^{13}N (9.965 min) and ^{11}C (20.39 min). After the short-lived ^{15}O was decayed, a radiation image of selected screens and the backup filter was obtained by 30-min exposure of one single IP (Size 35 x 43 cm). To evaluate contribution of ^{11}C and ^{13}N to the intensity of photostimulated luminescence (PSL), a separate air irradiation experiment was performed under the same irradiation and sampling conditions. After the irradiation the decay of activity for the screen and the backup filter was analyzed using GM detectors.

In comparison with ^{11}C, the 478-keV photopeak for ^{7}Be was analyzed using Ge detectors to obtain size distribution for ^{7}Be-bearing aerosol particles.
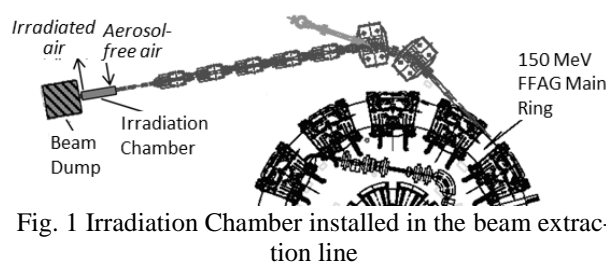

Fig. 1 Irradiation Chamber installed in the beam extraction line

RESULTS: The ^{11}C activities for each screen and the backup filter were estimated from the decay analysis mentioned above. The particle size of ^{13}N-bearing particles was not analyzed in this work because ^{13}N gaseous species like nitric acid interferes with collection of the ^{13}N-bearing particles [1]. The activity ratio of particles penetrating 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 [2]. Log-normal distributions are assumed as the size distribution function in the calculation because the number-based size distribution was confirmed to be log-normal by the SMPS measurement.

The geometric mean diameter, d_g, for the ^{11}C-bearing aerosol particles was successfully obtained using the combination way of the wire screen collection and the IP measurement. The diameter was found to be 68.0 nm for the ^{11}C-bearing particles, while that for ^{7}Be-bearing particles obtained by the Ge detector measurement was 63.4 nm. The activity-based geometric mean diameters were larger than the number-based one by ca. 20 nm. This shows that attachment process of radioactive atoms to the surface of aerosol particles is included in the radioactive particle formation.

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