PR2

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

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OBJECTIVE: Radioactive aerosols is one of the most important materials released in the severe accident of Fukushima Daiichi Nuclear Power Plant. The chemical and physical properties has been investigated for radioactive aerosols in the air and soil samples by various methods, and their properties have become cleared. On the other hand, their production mechanism in the reactor building has not been elucidated because the environment in the reactor buildings was anomalous: for example, high temperature, high dose rate, etc. In this project research, the production mechanism and measurement technique of radioactive aerosols have been studied experimentally to elucidate the generation mechanism in such particular environment. In addition, the behavior of radioactive aerosols after their deposition in the environment has been studied using simulated aerosols produced by radioactive aerosol generator developed in the present project.

RESEARCH SUBJECTS: The project research is composed by five individual subjects in this year period. One is developments of production method of radioactive aerosols for simulation experiments, one is development of measurement method of radioactive aerosols, and two are 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 the previous year period. The respective subjects of the research groups of this project are described as follows;

P2-1: Development of production method of radioactive aerosols by attaching radioisotopes to aerosol particles
P2-2: Development of measurement method of radioactive aerosols under severity conditions
P2-3: Investigation for production process of insoluble radioactive particles released from FDNPP
P2-4: Study for production mechanism of radioactive aerosols in the presence of sea water
P2-5: Interaction of radioactive cesium and suspended particles in river water

In this year period, experiments of subjects P2-3, -4 and -5 were not carried out because of limited operation of KUR.

RESULTS: In the subject P2-1, experiments using solution aerosol which was generated from three different sodium halide (sodium chloride, sodium bromide and sodium iodide) solution with various concentrations (0.005, 0.01 and 0.02 M) have been performed to elucidate the effect of solute on the attachment process of fission products to solution aerosol particles by the electrostatic interaction. The relationship between the attachment ratio of ^{104}Tc and total surface area of aerosol particles were obtained for each sodium halide solution aerosols of different concentrations. The equilibrium constant K was estimated by fitting to the relationship using adsorption equilibrium equation. In comparison of estimated K values among different solutes in the same concentration, it was found that the order of magnitude of K values is NaCl $\lesssim$ NaBr $\prec$ NaI. This order can be understood by the high anion concentration as compared with cation near the surface of aerosol particles as described in our previous report. On the other hand, in comparison of K values among the same solute in different concentrations, it seems that K values have no relation with the solute concentration. If the anion concentration near the surface of aerosol particles affect equilibrium constants, equilibrium constant could depend on the concentration. The reason of the independence might be caused by crystallization of solution aerosol particles. The anion concentration near the surface decreases if the crystallization occurred, which could induce decrease of K values. The concentration dependence has not been clear in the present research but will be elucidated by measurements using solutions of lower concentration in the future.

For the development of measurement method of radioactive aerosols under severity conditions (P2-2), an air irradiation experiment was performed in the 46-MeV electron linear accelerator facility of Institute for Integrated Radiation and Nuclear Science, Kyoto University. In the irradiation experiment, the performance of a newly assembled screen-type diffusion battery system consisting of three air-tight wire screen cylinders was examined. In the target room of the accelerator, a stainless steel irradiation tube was placed at a rear position of the tantalum target. During the irradiation, aerosol-free air was introduced to the tube from the experiment room next to the target room. The target was bombarded with a 30-MeV electron beam to produce bremsstrahlung and neutrons. The bremsstrahlung ionizes air and produces the radiation-induced aerosol. The particle size of ^{13}N-bearing aerosols formed in the accelerator was estimated using a combination technique of wire screens and imaging plate. Additionally the size distributions were measured by a scanning mobility particle sizer (SMPS) to compare the result with the developed system. Preliminary results showed that size of nano aerosol particles was in good agreement with that obtained with SMPS in the geometric mean diameter range of 10 to 15 nm. However, the size analysis for the ^{13}N-bearing particles was unsuccessful at the present stage because of low intensity of photostimulated luminescence of the imaging plate. It was mainly caused by subtraction of the gas condensation effect.

PR2-1 Effect of Solute on Attachment Behavior of Fission Product to Solution Aerosol Particle

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INTRODUCTION: Radioactive aerosol is one of transporting medium of radioactive materials, and transferred radioactive cesium and other radioactive materials from inside of the reactor building to the environment in the accident of Fukushima Daiichi Nuclear Power Plant. It was reported that the potential transport medium for radioactive cesium were sulfate aerosols[1]. But chemical form of radioactive cesium in the reactor building and attachment process to sulfate aerosol particles didn't become clear. The generation process of radioactive aerosols by attaching fission products to solution aerosol particles has been investigated experimentally by our research group. And it was found that there are two types of attachment processes; one is caused by geometric collision and another is induced by electrostatic interaction between a fission product and an aerosol particle. In the present work, experiments using solution aerosol which was generated from three different sodium halide solution with various concentrations have been performed to elucidate effect of solutes on the attachment process of fission products to solution aerosol particles by the electrostatic interaction.

EXPERIMENTS: The detail of the generation method of radioactive aerosols by attaching fission products to solution aerosol particles was shown in the previous reports [2, 3]. The fission products were released from a ^{252}Cf source in a chamber. The ^{252}Cf source was covered by a Havar foil of 50 μm thick to reduce the kinetic energy of fission products in the present experiments. Primary aerosols were generated from sodium halide solution (sodium chloride, sodium bromide and sodium iodide) of various concentration (0.005, 0.01 and 0.02 M) and injected into the chamber. The radioactive aerosol produced by attaching fission products to aerosol particles in the chamber were collected by a polycarbonate filter. On the other hand, all fission products released from the ^{252}Cf source into the chamber were collected by a grease-applied cellulose filter placed directly on the source. Gamma-ray spectrometry using a Ge-detector was performed for both filters to detect the gamma-ray emitted from the fission products, and the attachment ratio of fission products to aerosol particles was estimated from the ratio between fission product photo peak areas collected by both filters. In the present work, the attachment ratio for only ^{104}Tc could be estimated with enough statistics.

RESULTS: The relationship between the attachment ratio of 104Tc and total surface area of aerosol particles were obtained for solution aerosols of three different sodium halide (NaCl, NaBr and NaI) of three different concentrations (0.005, 0.01 and 0.02 M). The equilibrium constant K was estimated from the relationship as described in the previous report [4] and shown in Fig. 1. The upper, middle and lower panels indicate the relationship between equilibrium constants and concentration of solute for NaCl, NaBr and NaI, respectively. In comparison of K values among different solutes in the same concentration, it was found that the order of magnitude of K values is NaCl ≲ NaBr ≺ NaI. This order can be explained by the high anion concentration as compared with cation near the surface of aerosol particles [5] as described in the previous report [4]. And in comparison of K values among the same solute in different concentrations, K values increase from the concentration of 0.005 M to 0.01 M but decrease from that of 0.01 M to 0.02M. If the anion concentration near the surface of aerosol particles affect equilibrium constants, equilibrium constant could increase as concentration increase. The decrease of K values might be caused by crystallization of solution aerosol particles [6]. The anion concentration near the surface decreases if the crystallization occurred, which induces a decrease of K value. Even after taking into consideration the effect of the crystallization, the increase of K values from concentration of 0.05 M to 0.01 M could suggest that there is concentration effect on the attachment behavior of 104Tc to solution aerosol particles.

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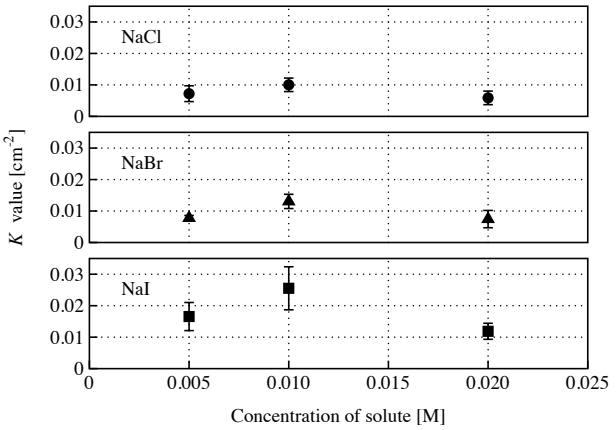

Fig. 1 Relationship between the equilibrium constant and concentration of solute.

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PR2-2 Particle Size Measurement of Radioactive Aerosol Particles in an Electron LINAC Using a Diffusion Battery System

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INTRODUCTION: Radiation-induced aerosol particles in the size range of several nm to ca. 100 nm 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 secondary radiation 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.

In this work, an air irradiation experiment was performed in the 46-MeV electron linear accelerator (LINAC) facility of the Research Reactor Institute (KURRI) to examine performance of a newly assembled screen-type diffusion battery system. We tried to analyze particle size distributions for both the whole radiation-induced aerosols particles and the particles bearing a radioactive atom.

EXPERIMENTS: The new diffusion battery (DB) system consisting of three air-tight wire screen cylinders was assembled. Each cylinder contained a stack of 500-mesh stainless steel wire screens, and it was connected to a ball valve and a mass flow controller to change a flow rate independently. The valve and mass flow controller were controlled by a PC for unmanned measurement of aerosol size distribution.

When very fine aerosol particles pass through a stack of wire screens, a part of the particles are trapped on 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 particle diameter 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: In the target room a stainless steel irradiation tube was placed at a rear position of the tantalum target. During the irradiation, aerosol-free air was introduced to the tube from the experiment room next to the target room. The target was bombarded with a 30-MeV electron beam to produce bremsstrahlung and neutrons. The bremsstrahlung ionizes air and produces the radiation-induced aerosol. The beam current was ca. 10-100 μA.

Size measurement for the whole radiation-induced aerosol particles: The irradiated air was simultaneously introduced to the two wire screen cylinders A and B. The air flow rate for Cylinder A was gradually changed from 0 to 15 L/min, while that for Cylinder B 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 tube. The irradiated air was sampled from both the upstream and downstream positions of the cylinders. The sampled air was introduced a condensation particle counter (CPC) to measure number concentration of the aerosol particles.

Size measurement for ^{13}N-bearing aerosol particles: In the previous work, particle size of ^{11}C-bearing aerosols formed in the FFAG proton accelerator of KURRI was estimated using a combination technique of wire screens and imaging plate (IP) [1]. The same technique was applied to the electron LINAC facility in this work. The irradiated air was introduced to Cylinders A and B at the same air flow rate. Each cylinder had 40 pieces of the 500-mesh screen and a PTFE backup filter. An additional PTFE filter was placed at the upstream of Cylinder B. In the electron LINAC, ^{13}N (half life: 9.965 min) is dominant radionuclide in air, and more than 90 % of ^{13}N exist as gas form. In such a circumstance, the ^{13}N gas often interfere the aerosol size measurement [2] due to condensation of the gas on the surface of the screens. Cylinder B was used estimation of the condensation by removing aerosol particles by the front PTFE filter. Activities of the screens and filters were simultaneously measured with a large IP (43 x 35 cm).

RESULTS: A scanning mobility particle sizer (SMPS) with nano-DMA was used for comparison with the DB system. The size distribution was confirmed to have a lognormal shape using SMPS in advance. The penetration ratios (N/N_0) obtained by changing air flow rate were fitted to a theoretical function [3] for lognormal distributions to obtain the geometric mean and geometric standard deviation of particle diameter.

Preliminary results showed that size of nano aerosol particles was in good agreement with that obtained with SMPS in the geometric mean diameter range of 10 to 15 nm. However, the size analysis for the ^{13}N-bearing particles was unsuccessful at the present stage because of low intensity of photostimulated luminescence (PSL) of the IP. It was mainly caused by subtraction of the gas condensation effect.

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