REENTRAINMENT OF $^{239}PuO_2$ PARTICLES CAPTURED ON HEPA FILTER FIBERS

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ABSTRACT

The collection efficiency of the air filter is the most important parameter for removing of radioactive aerosols. At the same time, retention of the captured particles is very important for contamination control not only during use but also after use. Reentrainment of $^{239}PuO_2$ particles collected on HEPA filter fibers was investigated under various conditions of air flow direction, air flow rate, air flow pattern and dust loading. Activity median aerodynamic diameter (AMAD) of Pu aerosols was about 0.45μm, and the geometric standard deviation (GSD) was 2. Collected activity on the filters ranged from 10^7 to 10^8 Bq/m^2. Dispersal rate under nominal direction flow was measured as less than 2 x 10^{-7} /hr. Under the inverse direction flow, higher dispersal rates were observed, but dispersal basically decreased with increasing sampling run. High air flow rate increased the dispersion. There was no significant differences among air flow patterns, continuous flow, intermittent flow and mechanical shock flow. Dispersal rate strongly depended on the dust loading. For example, three times loading of atmospheric airborne particles against the initial pressure drop was found to increase the dispersal rate by over 300 times to 10^2 /hr at maximum. Even if experimental conditions were considered extremely severe, it makes us to take reentrainment from a high loading filter more seriously, since it indicates that there is a possibility of air contamination due to handling of the spent HEPA filter.

INTRODUCTION

Reentrainment of particles collected by the air filter as well as a filtration mechanism has been a very important subject. The recoil phenomenon of alpha-emitting nuclides such as plutonium(Pu) has been recognized to affect retention of alpha active particulate on filters. There are many papers on the recoil of radon progeny from a substrate from the perspective of basic science and in practical application. However, there are few studies on the recoil of Pu particles. Fleischer and Raabe noted the possibility that recoiling fragments of Pu particles due to alpha decay relates to dissolution in liquids[1]. McDowell et al. investigated the effects of recoil on penetration through HEPA filters and reported that alpha-emitting particulate matter does indeed penetrate high efficiency filter media much more effectively than nonradioactive or beta-gamma-active aerosols[2),3]. However, Wen and Kasper investigated the kinetics of reentrainment from surfaces for nonradioactive particles and explained particle reentrainment as a result of a competition between adhesion forces and fluid dynamic lift forces[4].

In our previous studies[5),6] on HEPA filter, decontamination factors against Pu aerosol particles were almost the same or higher than the levels expected from experiments using nonradioactive aerosol particles, but these studies did not clarify retention efficiency. For filters in long-term use and spent filters containing such plutonium-containing dust, retention efficiency is very important along with collection efficiency. In this work, we focused on the retention of alpha active particulate, distinguishing from apparent retention with re-collection and net retention.

MATERIALS AND METHODS

Experiments were performed with filter media not in-place HEPA filters. The filters used in this work had an effective diameter of 37 mm. Basic collection performance was examined in our previous studies[6]. Two types of test filters with and without dust loading were prepared prior to loading of radioactivity for dispersion experiments. Atmospheric dust was loaded up to 1.7 and 3.0 times the initial pressure drops of the filter. Radioactive source filters were prepared by collecting plutonium oxide ($^{239}PuO_2$) aerosol particles[5] at an air flow face velocity of 7.8 mm/s, which corresponds to about one-third of the nominal flow velocity of HEPA filters. The particles were polydisperse aerosols with AMAD (Activity Median Aerodynamic Diameter) ranging from 0.4 to 0.5 μm and GSD (Geometric Standard Deviation) ranging from 1.9 to 2.1. Activity densities on the source filter surface ranged from 10^7 to 10^8 Bq/m^2.

Reentrainment of Pu particles captured on HEPA filter fibers was examined under dynamic conditions with air flow rather than static conditions. Source filters and clean filters for sampling of the dispersed particles were mounted in a multistage filter holder as shown in Fig.1. Experimental parameters on air flow through the source filter consisted of two air flow directions, two air flow velocities and three air flow patterns. The air flow directions were a nominal direction from front to back and an inverse direction from back to front. The air flow velocities were a nominal velocity of 23 mm/s and high velocity of 1.2 x 10^2 mm/s. The air flow patterns were (1) continuous flow, (2) intermittent flow that stopped for five seconds per half minute and (3) mechanical shock flow by free fall from a height of 50 mm. Three runs of five minutes sampling were repeated every ten days so that nine samples were obtained for each source filter.

SSD for alpha counting and phoswich detector for LX-rays of ^{239}Pu counting were used for activity measurements of filter samples. The minimal detectable activity during 2.5 x 10^5 sec counting was 1.2 x 10^{-2} Bq.

RESULTS AND DISCUSSION

Effects of air flow directions through the source filter on reentrainment of Pu particles were examined by intermittent flow at high air flow velocity. In the case of nominal direction flow, no significant activities in sampling filters were detected except one measurement among total 27 sampling runs. The dispersal rate per hour was very small and was estimated to be 2.3 x 10^{-7}. However, dispersed activities were observed in most runs with inverse flow from back to front. This indicates that particle reentrainment with nominal direction flow is not denied. Dispersed particles would be re-collected by fibers before penetrating a filter. The maximal dispersal rate per hour reached 2.4 x 10^{-5}. Dispersal rates scattered among source filter samples and even among successional runs using the same samples. Overall tendency showed a decreasing trend with the sampling runs. Next, effects of air flow patterns through the source filter on reentrainment of Pu particles were examined with inverse direction flow at high air flow velocity. Three source filter samples for each air flow pattern, continuous flow, intermittent flow and mechanical shock flow, were used for dispersion experiments. As shown in Fig.2, the measured dispersal rates ranged from 4 x 10^{-5} to 10^{-7} /hr. There were no differences in dispersal rates among air flow patterns. Thirdly, effects of air flow rates through the source filter on reentrainment of Pu particles were examined with intermittent flow in an inverse direction. The dispersal

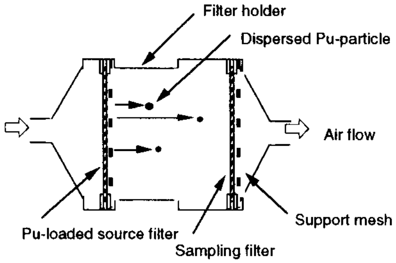

Fig.1 Structure of filter holder with multistage for sampling the dispersed Pu particles

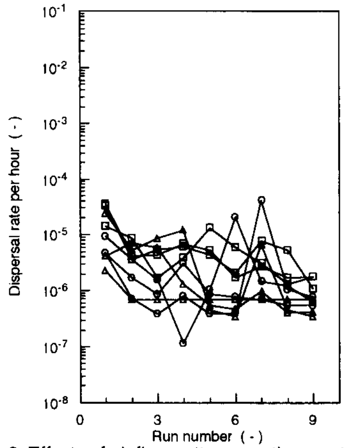

Fig.2 Effects of air flow patterns on the reentrainment of Pu particles at inverse flow of 1.2 x 10^2 mm/s (○: continuous flow, △: intermittent flow, □: mechanical shock flow)

rate depended on the air flow velocity. The measured dispersal rates at nominal velocity were smaller than those at high velocity. The maximal dispersal rate was 3.1 x 10^{-6} /hr and it was approximately one-tenth that at high velocity.

For source filters loaded with atmospheric aerosols, reentrainment was examined in the same way as for the pre-loaded source filters described above. The experiments were made in the inverse direction flow through the source filters at high flow velocity. Air flow patterns were continuous flow, intermittent flow and mechanical shock flow. As shown in Fig.3, high dispersal rates were observed for the source filters with dust loading. The maximal dispersal rates were 5.6 x 10^{-4} /hr with low dust loading and 1.0 x 10^{-2} /hr in high dust loading. These were 23 - 4.2 x 10^2 times higher than the rate for the filter without dust loading. There were no differences of the dispersal rates among the three air flow patterns. The dispersal rate decreased with increases in sampling run number. The dependency was

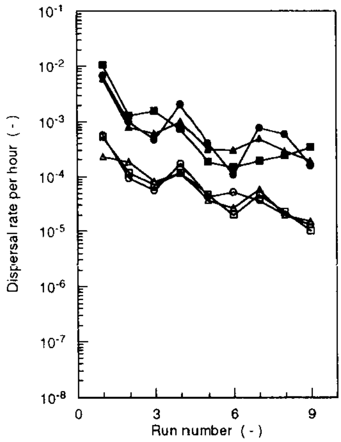

Fig.3 Effects of low and high dust loading on reentrainment of Pu particles at inverse flow of 1.2 x 10^2 mm/s (○: continuous flow, △: intermittent flow, □: mechanical shock flow, open symbol: low loading, solid symbol: high loading)

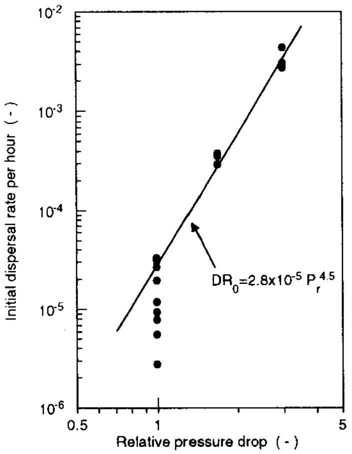

Fig.4 Dependency of the initial dispersal rate on the dust loading level of the source filter

relatively more clear than that for filters without dust loading. The fitted curves were expressed as an exponential function defined by

$$DR = a \exp(b N)$$

where DR is the dispersal rate per hour, N is the number of sampling runs, and a and b are the constants. b in the low and high loading filters were estimated as 0.36 and 0.34, respectively. The difference between these two was very small, so that factor b was assumed to be common (b = 0.35) for all experiments including those using filters without dust loading. Initial dispersal rate, DR_0, is defined by extrapolating to the point where the sampling run number is zero. The DR_0s for filters with low and high dust loading and those for filters without dust loading were estimated and plotted against the relative pressure drop, P_r, of the source filter to initial pressure, where the P_r is one of the indices indicating the dust loading levels. The DR_0 linearly increased with the P_r in a semi-logarithmic graph as shown in Fig.4. The relationship was expressed by

$$DR_0 = 2.8 \times 10^{-5} P_r^{4.5}.$$

For example, the dispersal rate at two times dust loading is calculated to have increased by 23 times. It was confirmed that the reentrainment was strongly affected by dust loading. The loaded dust particles play the role of a Pu carrier in radioactive particle reentrainment.

CONCLUSIONS

The reentrainment of submicron-sized Pu particles captured on HEPA filter fibers was experimentally investigated, and the results are summarized as follows:

(1) With nominal direction flow from front to back, the dispersal rate was estimated to be below 2.3 x 10^{-7} /hr at a high velocity of 1.2 x 10^2 mm/s. This dispersal rate was much lower than the maximal penetration of the HEPA filter with a minimal collection efficiency of 99.97 % for 0.3 μm sized particles. Therefore, it was concluded that Pu particles captured on filter fibers near the front surface hardly penetrate the filter.

(2) With inverse direction flow from back to front, reentrainment phenomena of Pu particles were observed in most experiments. The dispersal rate increased with air flow velocity and dust loading to the order of 1 x 10^{-2} /hr. In handling of a spent filter contaminated by Pu, marked attention should be paid to preventing air contamination.

(3) Dispersal rates were not constant in the succeeding sampling runs and decreased with large scattering. The fitted curves were expressed as an exponential function for all experiments.

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