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Attachment of ^{38}Cl and ^{39}Cl induced by high-energy neutrons to coexisted aerosols

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In order to confirm the effects of physicochemical properties such as liquid or solid of coexisted aerosol on radioactive aerosol formation, the number size and activity size distributions of ^{38}Cl and ^{39}Cl aerosols (just use Cl aerosols to protect confusion) generated by irradiation of argon (Ar) gas containing liquid dioctyl-phthalate (DOP) or solid NaCl aerosols with 45 MeV quasi-monoenergetic neutrons were measured. In the case of DOP aerosol, the ^{38}Cl and ^{39}Cl respectively formed from the (n, 2np) and (n, np) reactions of ^{40}Ar were observed. It was found that the activity size distribution curve shifted toward larger particle diameter, compared to the number size distribution. The measured activity size distribution of Cl aerosols was in good agreement with the calculated activity size distribution assuming the surface attachment of ^{38}Cl and ^{39}Cl to DOP aerosol. In the case of NaCl aerosol, the Cl aerosol formation could be also explained by the same attachment model in the case of DOP. These findings suggest that the formation mechanism of Cl aerosols is not affected by the physicochemical properties such as liquid or solid coexisted aerosols.

KEYWORDS: Radioactive aerosol formation, Attachment model, Size distribution, High-energy neutron, Internal dosimetry, High-energy accelerator

I. Introduction

In the air of accelerator and target rooms at high-energy proton accelerator facilities, radioactive aerosols or gases are produced[1-3]. The radiation protection against internal exposure due to the inhalation of the induced airborne radionuclides is one of the important problems at the accelerator facilities. For the assessment of internal doses due to the inhalation of airborne radionuclides, parameters such as a deposition rate in the respiratory tract should be determined. The dosimetric parameters are affected by the physicochemical properties such as particle sizes and chemical forms of airborne radionuclides. It is therefore important to clarify physicochemical properties, which has an influence on the formation of the radioactive aerosols.

The formation of radionuclides such as ^{7}Be, ^{11}C, ^{13}N, ^{15}O, ^{18}F, ^{24}Na, ^{38}S, ^{38}Cl and ^{39}Cl was observed in the air of an accelerator room and tunnel during the machine operation[4]. Endo et al. [2,5,6] reported the production rates, formation mechanisms and chemical forms of ^{11}C, ^{13}N and ^{15}O during the machine operation. Muramatsu[4] et al. showed that the geometric median radius of ^{24}Na, ^{7}Be and ^{38}S were about 28 nm. There is however very little information on the physicochemical properties and formation mechanism of airborne radiochlorine such as ^{38}Cl ($T_{1/2}$= 37.2 min) and ^{39}Cl ($T_{1/2}$= 55.6 min). Therefore, it is needed to clarify the chemical forms and formation mechanism of airborne radiochlorine.

In the previous reports[2, 7], the formation of non-radioactive coexisted aerosols such as sulfate and nitrate aerosols was observed in the air irradiated by the 12 GeV proton beam. These reports indicated that the radioactive aerosol formation could be explained by simple attachment reactions of radionuclides with the non-radioactive aerosol generated through radiation-induced reactions. Thus, size-distribution of radioactive aerosol is calculated on the basis of that of non-radioactive aerosol by using attachment model. But there are not only sulfate and nitrate aerosols but also many kinds of aerosol at accelerator facilities. Therefore, it was not clear whether an application of attachment model to all kinds of aerosols with different physicochemical property is possible or not. These indicate that the confirmation of physicochemical effects on attachment model is important. We have already reported that the existence of non-radioactive aerosol was necessary for the formation of ^{38}Cl and ^{39}Cl aerosol (just use Cl aerosol to protect confusion) from argon (Ar) gas due to the irradiation of 65 MeV neutron. It was also found that the formation mechanism of Cl aerosol can be explained by simple surface attachment of ^{38}Cl and ^{39}Cl to coexisted dioctyl-phthalate (DOP) aerosol[8,9].

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In the present study, DOP and NaCl aerosols as the representative aerosols with physicochemical properties such as liquid and solid have been selected to clarify whether an application of attachment model to different type aerosols in accelerator facilities is possible or not. Furthermore, the effects of physicochemical properties of coexisted aerosols on the formation mechanism of radioactive aerosols formed by high-energy neutron irradiation have been examined.

II. Experimental methods

The experiment was carried out using LC0 course in light-ion room 3 of TIARA (Takasaki Ion Accelerators for Advanced Radiation Application) of Japan Atomic Energy Research Institute. In the generation of 45 MeV quasi-monoenergetic neutrons, the ^{7}Li(p, n) reaction by bombarding a 3.80 mm thick ^{7}Li-target (99.8 %) with 50 MeV protons from an AVF cyclotron was employed. The generated neutron was transported to 10.9 cm-diameter iron collimator. Thereafter, the collimated neutron was introduced to the experimental room. **Figure 1** shows the diagram of the experimental setup. It consists of an atomizer (Model 3075, TSI Inc), an irradiation chamber (stainless steel) equipped with acrylic windows, an evaporation-condensation aerosol conditioner (Model 3072, TSI Inc.), a diffusion dryer, a scanning mobility particle sizer (SMPS) (Model 3936L22, TSI Inc) and an electrical low pressure impactor (ELPI) (ELPI-2000, DEKATI).

Firstly, the high purity (>99.995 %) Ar gas was introduced to the irradiation chamber, which was placed in the beam axis, at a flow rate of 20 L/min for 5 min. DOP or NaCl aerosols were produced by supplying solutions of 10% (V/V %) DOP in 2-propanol or 10% (W/V %) NaCl in water to the atomizer with Ar gas. The generated DOP aerosol was introduced to the evaporation-condensation aerosol conditioner in order to produce the lognormal distribution aerosols. In the case of NaCl aerosol, the diffusion dryer was used for dehydration.

The size distribution and particle number of were confirmed by SMPS before the introduction to the irradiation chamber. The measured median diameter of DOP and NaCl aerosols was 0.3 μm. Argon gas containing aerosol was introduced to the irradiation chamber at a flow rate of 1 L/min for 3 min. At the end of the introduction of aerosols, the concentrations of DOP or NaCl aerosols in the irradiation chamber were 7.8×10^{4} and 7.1×10^{3} particles/cm^{3}, respectively.

The prepared Ar gas containing aerosol was irradiated with the 45MeV quasi-monoenergetic neutron beam for 100 min. The spectrum of 45 MeV quasi-monoenergetic neutrons has been already reported by Baba et al[10]. The measurement of a neutron fluence rate was performed by same method as the previous papers[8,9]. A neutron fluence rate in the monoenergetic peak at the acrylic window of the irradiation chamber was 1.9×10^{4} cm$^{-2}\cdot$ s^{-1}.

After the irradiation, the irradiated aerosols were collected from the irradiation chamber at a flow rate of 10 L/min for 20 min and the number of collected aerosols on the each collection plates was measured using the electrometers of ELPI. To obtain the activity size distribution, the collection plates were removed from the impactor stages of ELPI after aerosol sampling and measurement of number size distribution, and the radioactivities of the collection plates were measured by a gas-flow counter. Then, γ-spectra of the collection plates were measured using a germanium semi-conductor detector. The decay correction was performed at the end of the irradiation for the quantification of a production rate due to nuclear reactions.

III. Results and Discussion

Figure 2 shows the typical γ-ray spectrum of the collection plates of ELPI in the case of NaCl aerosol addition. The ^{38}Cl and ^{39}Cl respectively formed from the (n, 2np) and (n, np) reactions of ^{40}Ar were observed. The ^{38}Cl and ^{39}Cl were also found in the case of DOP aerosol (data not shown).

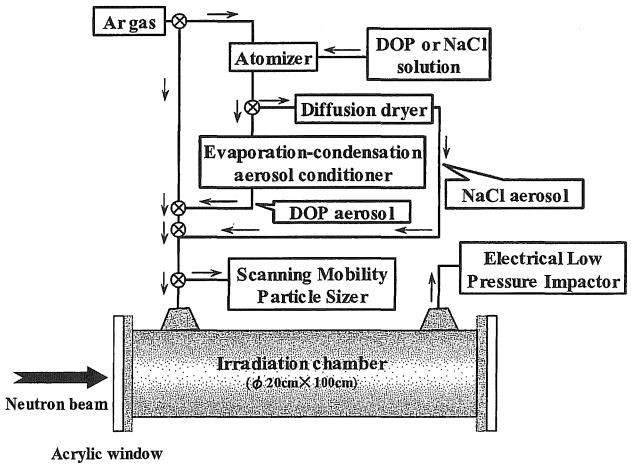

Fig. 1 Diagram of the experimental setup

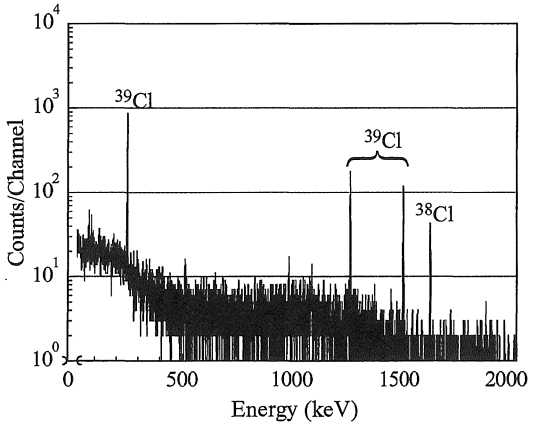

Fig. 2 Typical γ-ray spectrum of the collection plates of ELPI after the irradiation of 45 MeV neutron in the case of NaCl aerosol.

The number size and activity size distributions of Cl aerosols were analyzed on the basis of simple attachment model according to a method proposed by Porstendörfer[11,12,13]. The calculation of attachment coefficient was performed using average mass of ^{38}Cl and ^{39}Cl, since it was estimated from the calculation using the IRACM code[14] that the amount of produced ^{38}Cl is almost same as that of ^{39}Cl.

Figure 3 shows the number and activity size distribution of ^{38}Cl attached liquid DOP aerosol. The distribution curves were normalized to the respective maximum concentrations. Like the results of the previous experiments[8,9], the measured activity size distribution of Cl aerosols shifted toward larger particle diameter, compared to the number size distribution (**Fig. 3**(a)). The geometrical median diameters (GMD) of fitted number size and fitted activity size distribution were 0.45 and 0.59 μm, respectively. The fitted activity size distribution of Cl aerosols was in good agreement with the calculated activity size distribution (GMD = 0.43 μm) assuming the attachment of ^{38}Cl and ^{39}Cl to DOP aerosols (**Fig. 3**(b)).

To examine the influence of physicochemical property of coexisted aerosol to the attachment behavior, NaCl solid aerosol was used as coexisted aerosol instead of DOP liquid aerosol (**Fig. 4**). Like the case of DOP aerosols, the fitted activity size distribution of Cl aerosol was greater than that of fitted number size distribution. The GMDs of number and activity size distributions were 0.46 and 0.66 μm, respectively (**Fig. 4**(b)). The fitted activity size distribution agreed with the computed activity size distribution (GMD = 0.44 μm), meaning that the formation of Cl aerosols could be explained by the attachment model[11] as well as the case of DOP aerosols. These results indicates that the application of the attachment model to coexisted aerosols with physicochemical properties is possible.

In the previous papers[2,4,15,16,17], it has been already suggested that non-radioactive aerosols are mainly produced by radiation induced oxidation reactions of SO_2 present in the air of high-energy accelerator facilities and a physicochemical type of produced aerosols is liquid type. These reports indicate that the formation of ^{24}Na, ^{7}Be and ^{38}S aerosols is simple attachment reaction. As shown in **Fig. 3** and **Fig. 4**, the present study showed that the formation mechanism of Cl aerosols formed by high-energy neutron was not affected by the physicochemical properties such as liquid or solid. The finding indicate that the physicochemical properties such as liquid or solid of coexisted aerosols do not affect the

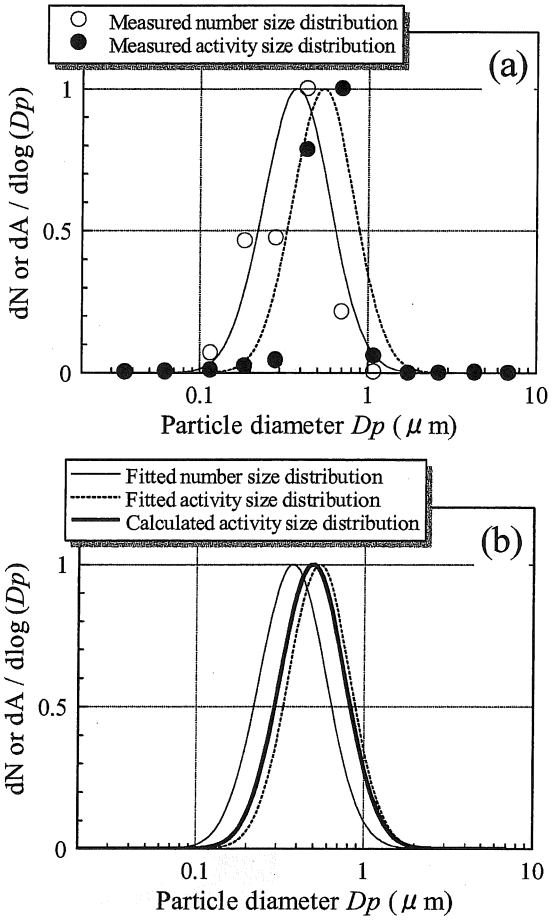

Fig. 3 Size distribution of ^{38}Cl and ^{39}Cl attached DOP aerosols formed by the irradiation of 45MeV neutrons, (a) measured number size and activity size distributions, (b) fitted number size, fitted activity size and calculated activity size distribution.

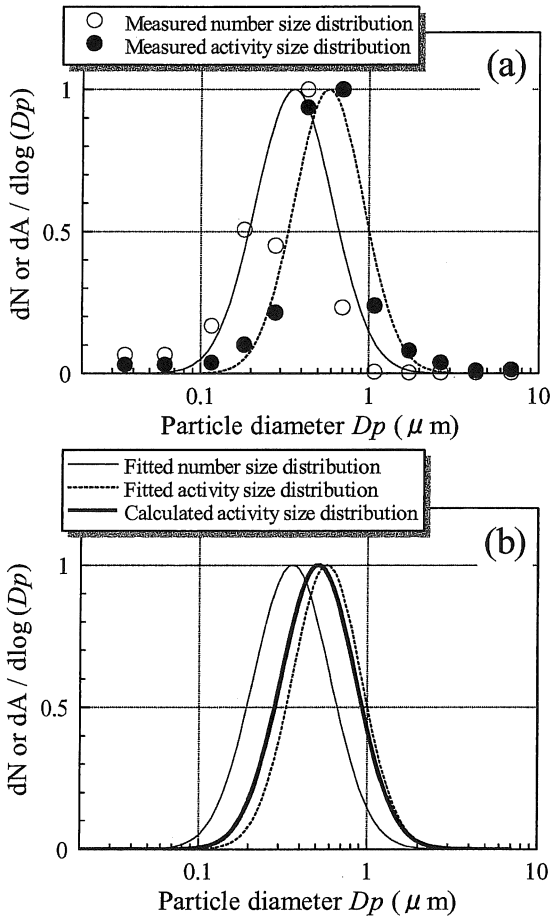

Fig. 4 Size distribution of ^{38}Cl and ^{39}Cl attached NaCl aerosols formed by the irradiation of 45MeV neutrons. Notation in the figure are the same as those in **Fig. 3**.

formation of the Cl aerosols induced due to an irradiation of high-energy neutron to Ar gas.

IV. Summry

The effects of physicochemical properties of coexisted aerosols on the formation mechanism of Cl aerosols were examined. When NaCl aerosol instead of DOP aerosol was coexisted aerosol, the formation of ^{38}Cl and ^{39}Cl attached NaCl aerosol can be explained by the same attachment model as the DOP aerosol.

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