Particle Size of Radioactive Aerosols Generated During Machine Operation in High-energy Proton Accelerators

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INTRODUCTION

Radioactive nuclides are formed through nuclear reactions in the air of accelerator tunnels during machine operation. Especially, in high-energy accelerator facilities, various radionuclides are produced from the air through many kinds of nuclear reactions, such as nuclear spallation reactions. In addition, non-radioactive aerosols are abundantly generated in the air due to high radiation doses in the operating accelerators. Under such a condition, the produced radioactive atoms form radioactive aerosols. These aerosols might be inhaled by workers who enter the tunnel just after beam stoppage. The particle size is very important information both for estimating the internal exposure doses and for designing the exhaust equipment of accelerator facilities. We installed a flow-type chamber in the slow-extracted proton beam line (EP2) of the 12-GeV proton synchrotron at High Energy Accelerator Research Organization (KEK) in order to intensively investigate the characteristics of the radioactive airborne species. The chamber makes it possible to precisely analyze radioactive aerosols formed by the irradiation of high-energy particles. Air can be supplied to the chamber under controlled conditions, and returning irradiated air is analyzed concerning the basic properties of aerosols observed in the air.

In this work, focusing on typical radionuclides produced in the tunnel air, such as ¹¹C, ¹³N, ⁷Be and ²⁴Na, their aerosol size distributions are reported. In addition, the internal doses due to inhalation of radioactive aerosols were estimated using the obtained parameters for the aerosol size.

EXPERIMENTAL

(a) Sampling of aerosols

Figure 1 shows a plane view of the EP2 beam line of KEK. In the EP2 line, a platinum target (1cm dia, 6cm long) was used in the primary proton beam line to produce secondary particles, such as K, π-mesons. A flow-type chamber was placed between the Pt target and an iron beam dump. The chamber was made of aluminum and its volume was 0.6 m³. A measurement station was placed outside of the tunnel, as shown in Fig. 1. The chamber and the station were connected with a supply and return tubes. They were made of stainless steel, and were 20 m long and 5.4 cm in diameter.

Figure 2 shows the measurement station. Aerosol-free air was prepared by filtering air with a HEPA filter, and was supplied from the measurement station to the chamber during machine operation at a flow rate of ~4 m³/h. The air irradiated in the chamber was returned to the measurement station. The temperature and relative humidity of the returned air were 296 K and 42%, respectively. The intensity of the 12-GeV primary proton beams was approximately 3.8 x 10¹ⁱ particles per sec, which was monitored with a secondary emission chamber (SEC). The exposure rate around the chamber was 10⁵ ~ 10⁷ R/h during machine operation.

Fig. 1 A plane view of the EP2 beam line
(b) Determination of the particle-size distribution

In this work, a screen-type diffusion battery was employed in the aerosol-size analysis. The battery consisted of stacked wire screens, whose number could be changed. The screen was made of thin stainless-steel wires (500 mesh), and was 4 cm in diameter. The aerosols in air decrease in number when the air is passed through the diffusion battery. The relation between the concentration of aerosols at the inlet and the exit of the battery was derived theoretically by Cheng and Yeh (2). Assuming that the aerosols have log-normal size distributions, the relation can be expressed as a function of the number of screens, the diffusion coefficient of the aerosols, and the flow rate of air (3),

\[ \frac{N}{N_0} = \frac{1}{\sqrt{2\pi} \log \sigma_g} \int_{-\infty}^{\infty} \exp \left\{ -KN_S \left( \frac{Q}{D(r)} \right)^{\frac{3}{2}} \right\} \exp \left\{ -\frac{(\log r - \log r_g)^2}{2\log^2 \sigma_g} \right\} d\log r, \]

where, \( N_0 \) and \( N \) are the particle concentrations at the entrance and exit of the diffusion battery, \( r \) is the particle radius of the aerosols, \( r_g \) and \( \sigma_g \) are the geometric mean and geometric standard deviation of the particle radius, \( Q \) is the flow rate of the sample air, \( D(r) \) is the diffusion coefficient, and \( K \) is a specific constant for the screen used (\( K=254 \)).

The diffusion constant \( D(r) \), which is a function of the particle radius, is represented as

\[ D(r) = \frac{\kappa T}{6\pi \eta r} \left[ 1 + A \frac{L}{r} + C \frac{L}{r} \exp \left( -\frac{Br}{L} \right) \right], \]

where, \( A, B \) and \( C \) are the coefficients for Cunningham's correction \( (A = 1.246, B = 0.87 \text{ and } C = 0.42 \text{ (1)}) \), \( \kappa \) is Boltzmann's constant \( (1.38 \times 10^{-16} \text{ erg K}^{-1}) \), \( T \) is the absolute temperature \( (K) \), \( L \) is the mean-free path of air molecules \( (0.653 \times 10^{-5} \text{ cm}) \), and \( \eta \) is the viscosity of the air \( (1.83 \times 10^{-4} \text{ dyne cm}^{-2} \text{ at } 296 \text{ K and } 101.3 \text{ kPa}) \).

From equations (1) and (2), \( r_g \) and \( \sigma_g \) can be derived using the values of the penetration fraction \( (N/N_0) \), which is obtained experimentally.

The penetration fraction was measured both for non-radioactive and for radioactive aerosols. For non-radioactive aerosols, their concentrations, \( N_0 \) and \( N \), were measured using a condensation particle counter ((CPC); TSI, Inc., Model 3025A), as shown in Fig. 2. For radioactive aerosols, i.e. aerosols bearing radioactive nuclide, \( N/N_0 \) can be replaced by \( A/A_0 \), which corresponds to the ratio of the radioactivity of the nuclides \( (A) \) which passed through the diffusion battery to the total radioactivity \( (A_0) \).

Two diffusion batteries were employed in this experiment. One was that for \( N_0 \) and \( A_0 \) with a single screen \( (D_1 \text{ in Fig. 2}) \), and the other \( (D_n) \) was that for \( N \) and \( A \) with plural screens whose number, \( n \), could be changed. The screen in \( D_1 \) and the first screen in \( D_n \) were used to remove the fraction of free radioactive atoms with large diffusivity. The obtained penetration fractions were plotted vs \( n - 1 \). A part of the returned air
from the chamber was simultaneously introduced to both diffusion batteries (as shown in Fig. 2) at the same flow rate of 13.0 L/min. The flow rate was precisely adjusted with mass-flow controllers. The aerosols which passed through the batteries were collected for 20 min on membrane filters (Millipore Co. Type HA) of 47 mm diameter with a pore size of 0.45 µm; the collection efficiency exceeds 99% for the present sampling conditions.

(c) Radioactivity measurement
The activity collected on the filters was measured with a high-resolution Ge semiconductor detector connected to a 4k pulse-height analyzer. The activity was determined using a $^{152}$Eu standard source with the same counting geometry as that of the sample filters. The photopeaks used were the 478-keV for $^7$Be and the 1368-keV for $^{24}$Na. The activities for $^{11}$C and $^{13}$N were determined by a least-squares fitting of the decay curve of the 511-keV photopeak. The concentration of radioactive aerosols was calculated from the activity by correcting the radioactive decay during and after the sampling.

RESULTS AND DISCUSSION
(a) Radioactive aerosols formed in the chamber
Various radionuclides were produced through nuclear reactions from air passing through the chamber during machine operation. Figure 3 shows the $\gamma$-spectrum of the aerosols that were collected on a filter without passing through the diffusion battery. The spectrum was measured for 1 min 8 min after the end of the aerosol sampling. The observed radionuclides were $^7$Be (half life: 53.3 d), $^{24}$Na (15.0 h), $^{27}$Mg (9.46 m), $^{28}$Al (2.24 m), $^{29}$Al (6.6 m), $^{38}$S (5.0 m), $^{39}$Si (2.87 h), $^{39}$Cl (37.2 m), $^{39}$Cl (56.2 m). A 511-keV peak for annihilation $\gamma$-rays was also observed strongly. The 511-keV $\gamma$-rays are emitted from positron emitters such as $^{15}$O (half life: 2.037 min), $^{11}$C (20.39 min), $^{13}$N (9.965 min), $^{18}$F (109.8 min). The activity was drastically decreased when a mixture of pure nitrogen and oxygen (N$_2$ 79 %, O$_2$ 21 %) was passed through the chamber instead of the filtered air. This result indicates that most of radioactivities, except for $^{15}$O, $^{11}$C, $^{13}$N, $^{18}$F, are produced through nuclear spallation reactions from argon in air.

![Fig.3 Typical $\gamma$-spectrum of the collected aerosols](image)

The number of non-radioactive aerosols, counted with the CPC, was found to be $\sim 4 \times 10^6$ /cm$^3$. It depends on the beam intensity and the flow rate of the supplied air. The formation of a large number of fine aerosols was a characteristic phenomenon observed in the tunnel of operating accelerators where high-radiation fields were generated. Kondo et al. previously suggested that most of the aerosols in the accelerator tunnel air were sub-micron, which were produced through radiation-induced reactions during machine operation; their main components have been identified as sulphate aerosols (4,5).

Figure 4 shows the relationship between the count of 511-keV photopeak of the filter and the elapse of time after the end of the sampling. The photopeak consists of four positron emitters, i.e., $^{15}$O, $^{11}$C, $^{13}$N, $^{18}$F. The decay curve was fitted into the four components by a least-squares method. The concentrations of the radioactive aerosols, which were obtained by correcting for decay during and after the sampling, were $5.4 \times 10^2$ Bq/cm$^3$ for $^{11}$C, $6.3 \times 10^2$ Bq/cm$^3$ for $^{13}$N, $3.3 \times 10^4$ Bq/cm$^3$ for $^7$Be and $2.4 \times 10^4$ Bq/cm$^3$ for $^{24}$Na.
(b) Particle size distribution for the aerosols

Figure 5 shows the relation between the penetration fraction \( \frac{N}{N_0} \) or \( \frac{A}{A_0} \) and the number of screens. The geometric mean \( r_g \) and standard deviation \( \sigma_g \) of the size distributions were obtained by a least-squares fitting from equations (1) and (2). The solid lines represent the results of a best fit.

Table 1 shows the geometric mean of the particle radius \( r_g \) and the geometric standard deviation \( \sigma_g \) which were obtained by the fitting. The size distributions for \(^{15}\text{O}\) and \(^{18}\text{F}\) were not analyzed because precise results could not be obtained due to its short half life for \(^{15}\text{O}\) and the low activity for \(^{18}\text{F}\). The mean radius ranged from 0.03 \( \mu \text{m} \) to 0.05 \( \mu \text{m} \), which was much smaller than that of ordinary atmospheric aerosols.

<table>
<thead>
<tr>
<th>Element</th>
<th>( r_g ) (( \mu \text{m} ))</th>
<th>( \sigma_g )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Non-radioactive</td>
<td>0.028</td>
<td>2.2</td>
</tr>
<tr>
<td>(^{11}\text{C})</td>
<td>0.035</td>
<td>1.8</td>
</tr>
<tr>
<td>(^{13}\text{N})</td>
<td>0.046</td>
<td>1.8</td>
</tr>
<tr>
<td>(^{7}\text{Be})</td>
<td>0.04</td>
<td>1.9</td>
</tr>
<tr>
<td>(^{24}\text{Na})</td>
<td>0.039</td>
<td>2.0</td>
</tr>
</tbody>
</table>

It was shown that the radioactive aerosols were larger than the non-radioactive aerosols. In our previous works, the particle size of radioactive aerosols was also studied both for radionuclides formed through nuclear reactions such as \(^{7}\text{Be}\), \(^{24}\text{Na}\), \(^{38}\text{S}\) (4,5) and for a natural radionuclide, \(^{218}\text{Po}\) (8), which is a daughter of radon emitted from the concrete wall of the accelerator tunnel. The experiments were carried out in the same EP2.
beam line under almost the same beam conditions. The size of radioactive aerosols was also found to be larger than that of non-radioactive ones in previous studies (4,5,8). In general, the key process for the formation of radioactive aerosols is the attachment of radioactive atoms to non-radioactive aerosol particles. If radioactive aerosols are formed through this simple attachment process, the size of the radioactive aerosol can be expressed by multiplying the size distribution for non-radioactive aerosols \( N(r) \) by the attachment coefficient \( \beta(r) \). \( \beta(r) \) was calculated (7) from the following equation, assuming that these radioactive atoms are neutral and that the attachment proceeds via simple physical adsorption reactions:

\[
\beta(r) = \frac{\pi r^2 \nu}{1 + \frac{\nu}{D}}, \tag{3}
\]

where \( r \) is the aerosol radius, \( \nu \) the average velocity and \( D \) the diffusion coefficient; \( \nu \) and \( D \) are defined as

\[
\nu = \sqrt{\frac{8kT}{\pi M}},
\]

\[
D = \frac{3L}{8} \sqrt{\frac{\pi kT m + M}{mM}},
\]

where, \( m \) and \( M \) are the mass of radionuclide and air molecule, \( T \) the absolute temperature, \( \kappa \) Boltzmann's constant and \( L \) the mean free path of air molecules.

In the previous work, the size distribution for the radioactive aerosols agreed well with the distribution calculated from the equation (3). However, the size of radioactive aerosols in the present work was slightly smaller than the calculated one. This result may be attributed to the large difference of the air-sampling rate from the tunnel between the present and previous studies. In the present work, the air returned from the chamber at the flow rate of 4 m\(^3\)/h was analyzed, while air was sampled directly from the tunnel at a flow rate of \( \approx 70 \) m\(^3\)/h for the aerosol size analysis in the previous studies. In general, aerosols grow through a coagulation process of aerosols. When air was sampled from the tunnel at a slow rate, the size distribution for radioactive aerosols could not be simply explained using the above-mentioned attachment model because other processes, like coagulation, proceed simultaneously with the attachment process of the radioactive atoms during the passing time of air through the chamber. The formation mechanism will be discussed elsewhere in detail.

(c) Internal doses due to the inhalation of radioactive aerosols

The dose coefficients for the inhalation of radioactive aerosols were estimated for \(^{11}\text{C}\), \(^{13}\text{N}\), \(^{7}\text{Be}\) and \(^{24}\text{Na}\), based on the respiratory-tract model of ICRP Publication 66 (9). They were calculated with the computer program LUDEP (10) using the geometric mean and the standard deviation of the aerosol size obtained in this work. In the calculation, the breathing rate of workers was assumed to be 1.2 m\(^3\)/h for a reference man of light work. The biokinetic models for elements given in ICRP Publication 30 (11) were applied to the calculation. The model for hydrogen was used for nitrogen because no model for nitrogen was provided in ICRP Publication 30. The calculated coefficients for the absorption type M were 1.1 \( \times 10^{11} \) Sv Bq\(^{-1}\) for \(^{11}\text{C}\), 6.2 \( \times 10^{12} \) Sv Bq\(^{-1}\) for \(^{13}\text{N}\), 1.4 \( \times 10^{10} \) Sv Bq\(^{-1}\) for \(^{7}\text{Be}\) and 5.7 \( \times 10^{10} \) Sv Bq\(^{-1}\) for \(^{24}\text{Na}\). Figure 6 depicts the internal dose rates as a function of time after beam stoppage. This relation was obtained from the above-mentioned concentration of the radionuclides and the dose coefficients. The \(^{11}\text{C}\) aerosols contribute mostly to the internal doses up to about 40 min after beam stoppage.

CONCLUSIONS

The aerosol sizes of the radioactive and non-radioactive aerosols were analyzed in the high-energy proton accelerator using diffusion battery. The aerosol radius was found to be 0.035 \( \mu m \) for \(^{11}\text{C}\), 0.046 \( \mu m \) for \(^{13}\text{N}\), 0.04 \( \mu m \) for \(^{7}\text{Be}\), 0.039 \( \mu m \) for \(^{24}\text{Na}\) and 0.028 \( \mu m \) for non-radioactive aerosols. The \(^{11}\text{C}\) aerosols give the largest contribution to the internal dose.
REFERENCES