PARTICLE SIZE DISTRIBUTION OF RADIOACTIVE AEROSOLS FORMED IN HIGH ENERGY ACCELERATOR TUNNELS

H. Muramatsu, K. Kondo, Y. Kanda and S. Takahara

National Laboratory for High Energy Physics
Oho-machi, Tsukuba-gun, Ibaraki-ken, 305, Japan

ABSTRACT

The ^{38}S, ^{24}Na and ^{7}Be, produced by nuclear spallation reactions in the air of high energy accelerator tunnels, are present in the form of an aerosol. The size distributions of these aerosols were measured with a parallel plate diffusion battery and found to be a log-normal distribution with a geometric mean radii of about 0.03 μm and a logarithmic standard deviation of 0.3.

INTRODUCTION

Many radioactive airbornes have been observed in the air of high energy accelerator tunnels during machine operation, and a part of them is present as radioactive aerosols.[1,2,3] However thus far little is known of their size distribution and the mechanisms of aerosol formation. The informations on their particle size distributions are very important in evaluating internal radiation doses of accelerator maintenance workers especially for the works right after machine shut-down.

In this paper, the aerosol size distributions of ^{38}S, ^{24}Na and ^{7}Be were examined extensively by using a parallel plate diffusion battery at the slow extracted proton beam line (EP2) of te National Laboratory for High Energy Physics (KEK). And the mechanisms of aerosol formation were discussed on the basis of an attachment reaction of these free radioactive atoms with ambient non-radioactive aerosols, which are mainly formed by radiation induced reactions.

EXPERIMENTAL

Fig. 1-A shows the plain view of the slow extracted proton beam line (EP2) of the KEK 12-GeV proton synchrotron, at which the air was sampled during machine operation. As Fig. 1-B shows, the air sampling inlet is about 50 cm above the beam axis and ~ 3.5 m downstream from the K2 target. The air sampled was introduced to te measurement station through a polyethylene pipe 5 cm in diameter and returned into the tunnel after measurement. Fig. 2 shows a schematic diagram of the measurement. The tunnel (~ 1400 m^3 in volume) was air-conditioned and ventilated at a rate of ~700m^3/h, and temperature and relative humidity were 295.4K and 39.4% respectively. The primary proton beam intensity was monitored with a SEC (secondary emission chamber) and was approximately 1.7×10^{11} protons/s.

The filters used for the collection of aerosols were membrane filters (Millipore Co. Type HA) of 47 mm in diameter with a pore size of 0.45 μm and/or 0.8 μm, the collection efficiency was over 99% for the present sampling conditions. A flow rate correction was applied by measuring the pressure differences with Hg manometers. The radioactivities of the filters were measured with a high resolution Ge(Li)-detector connected to a 4K PHA.

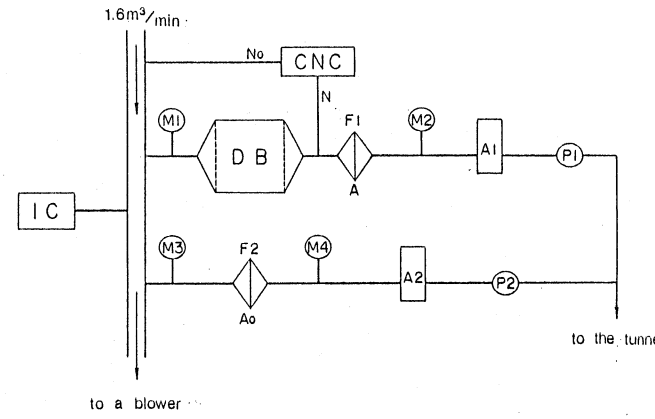

Fig. 2 Shematic diagram of measurements
CNC: condensation nuclei counter, DB: parallel plate diffusion battery, F1, F2: filter, A1,A2: flow-meter, P1,P2: air pump, M1 ~ M4: mercury manometer, IC: ionization chamber, A, A_0 : radioactivities collected on the filters, N, N_0 : aerosol numbers

RESULTS AND DISCUSSION

The gamma-ray spectra of radioactivities collected on the filter are shown in Fig. 3. More than 99% of ^{7}Be, ^{22}Na, ^{24}Na and ^{28}Mg are present in the form of aerosols, while short-lived isotopes such as ^{11}C, ^{15}O, ^{13}N and ^{18}F are mostly gaseous and the concentrations are of the order of $10^{4} \sim 10^{6}$ Bq/m^3 .

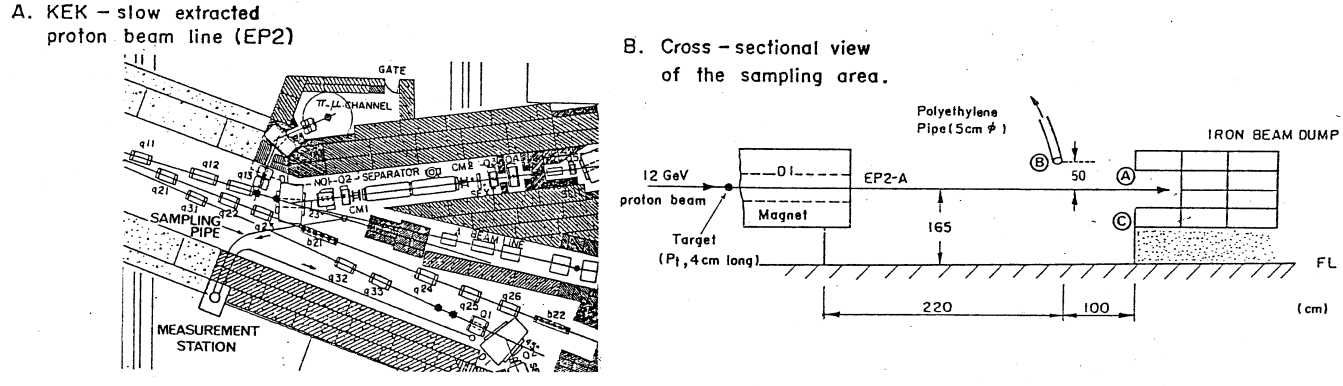

Fig. 1 Plain and cross-sectional views around the sampling inlet at the slow extracted proton beam line.

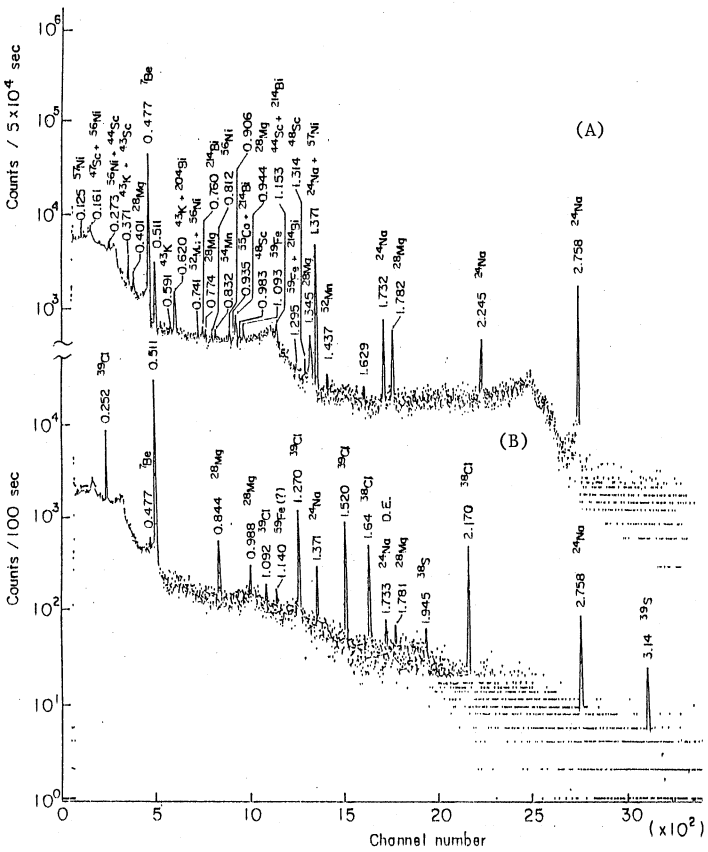

Fig. 3 Gamma-spectra of the radioactivity collected on the filter.
Cooling time: (A) 26.5 hours
(B) 12 min.

The diffusion battery with parallel-sided channels was used to determine aerosol size distributions. Generally aerosols are polydisperse and have a log-normal distribution. For a log-normal distribution of particle sizes, the penetration fraction (P) is given by the generalized DeMarcus' equation[4].

$$P = \frac{n}{n_0} = \frac{0.9149}{\sqrt{2\pi}\,\log\sigma_g}\int_{-\infty}^{+\infty}\exp(-3.77\,)\cdot\exp\left[\frac{-(\log r-\log r_g)^2}{2\,\log^2\sigma_g}\right]d\log r$$

$$+\frac{0.0592}{\sqrt{2\pi}\,\log\sigma_g}\int_{-\infty}^{+\infty}\exp(-44.66\,)\cdot\exp\left[\frac{-(\log r-\log r_g)^2}{2\,\log^2\sigma_g}\right]d\log r \qquad (1)$$

where r_g and σ_g are mean geometric radius and standard geometric deviation respectively, and n and n_0 are the particle concentrations at the exit and entrance of the diffusion battery. ν and diffusion coefficient(D) are defined by

$$\nu = \frac{NDb\ell}{tQ}\,,\quad D = \frac{kT}{6\pi\eta r}\left(1 + A\frac{L}{r} + C\frac{L}{r}\,e^{-Br/L}\right)$$

where N is the channel number of a diffusion battery, r the aerosol radius, t the half width between plates, b the height of the battery, ℓ the length of the battery, Q the flow rate, k Boltzmann's const. (1.38×10^{6} erg. K^{-1}), T the absolute temperature (K), L the mean free path of air molecules (0.635×10^{-5} cm), and η the viscosity of the air (1.83×10^{-4} dyne · cm^{-2} at 296 K and 101.3 KPa), A, B and C are coefficients for Cunningham's correction (A = 1.246, B = 0.87, C = 0.42)[10].

The dimensions of the diffusion battery used in this experiment are N = 20, t = 0.05 cm, b = 20 cm and ℓ = 57 cm respectively. From the relation of P vs Q experimentally obtained, r_g and σ_g were determined by using the SALS[5] computer program, which includes a least-square fitting method. In determing the radioactive aerosol distribution, n/n_0 can be replaced by A/A_0, which corresponds to the radioactivity ratio. The relation of A/A_0 vs Q experimentally obtained are shown in Fig. 4. The experimental erros in the figure are the standard deviations for the counting statics. The solid line computed as te best fit to the experimental data shows that ^{7}Be, ^{22}Na, ^{24}Na and ^{28}Mg have similar aerosol distributions with a geometric mean radius of r 0.028 μm and log $\sigma g \approx 0.35$. The log-normal distribution of these aerosols thus obtained is shown in Fig. 5.

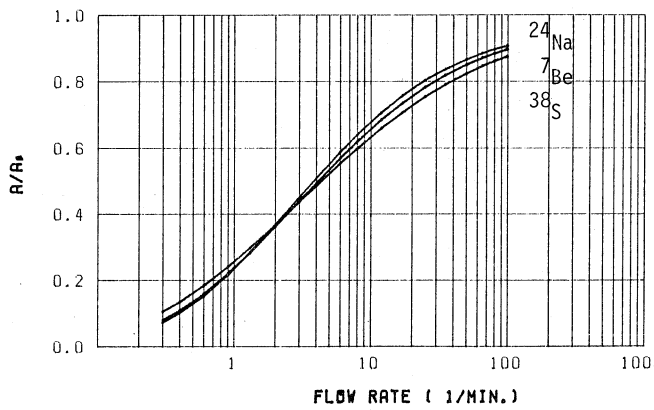

Fig. 4 The relation of A/A_0 vs Q

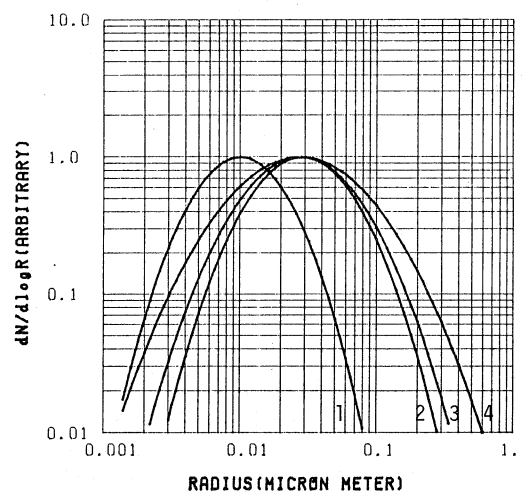

Fig. 5 The log-normal distribution of ^{38}S, ^{22}Na, and ^{7}Be aerosols
1: non-radioactive aerosol
2: ^{24}Na,
3: ^{7}Be
4: ^{38}S

On the other hand, the non-radioactive aerosol mainly produced by radiation induced reactions was measured at the same time and was of a log-normal shape with a geometric mean radius of 0.01 μm and $\log \sigma_g$ = 0.3, whereas the aerosol concentrations were (1.5~ 4.3)× $10^5/cm^3$, depending upon the beam intensity.

According to the attachment mechanism, the radioactive aerosol size distribution (F) has the following relation with a non-radioactive aerosol size distribution (N(r)):

$$F \propto \beta(r) \cdot N(r)$$

where $\beta(r)$ is an attachment coefficient determined by using the equation introduced by Natanson[6] and Lassen and Rau[7] . The Fig.5 shows the curve of $\beta(r) \cdot N(r)$ thus calculated, by assuming 15 % of radioactive atoms are charged.

REFERENCES

1. "Accelerator Health Physics", (Edited by H.W. Patterson and R.H. Thomas), p.524, 1973, Academic Press, New York and London.
2. S. Charalambus, A. Rindi, Nucl. Instrum. Meth. 56 (1967) 125.
3. K. Kondo, H. Muramatsu, Y. Kanda, S. Takahara, to be published in Int. J. appl. Radiat. Isotopes.
4. W. DeMarcus, J. Thomas, ORNL-1431 (1952).
5. The SALS program was developed by T. Nakagawa and Y. Oyanagi of Tokyo University, 1978.
6. G.L.J. Natanson, Tech. Phys. 30 (1978) 573.
7. L. Lassen, G.L.Rau, Z. Phys., 160 (1960) 504.