

matically analyzed on a computer, to provide the cloud-cover distribution as a function of time for use in studies of atmospheric dynamics and long-term changes in climate.

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## Fibrous Filters as Particle-Size Analyzers

**Abstract.** A method was developed for characterizing radioactive aerosols by determining their distribution as a function of depth in fibrous filters under carefully controlled conditions. The method distinguishes the contributions of three major processes of filtration: diffusion, interception, and inertial impaction. For those experiments where diffusion is the controlling mechanism, particle sizes in the range of 50 to 500 Å thus determined compare favorably with those obtained from concentration and flow data and electron photomicrographs.

The characterization of gas-borne particulate material, especially fission products which are released during a reactor accident, is necessary so that effective means may be developed for removing radioactivity from gas streams.

We have developed a method for determining the sizes of radioactive particles, based on their interaction with fibers, using special techniques, considerably refined over the one first suggested by Sisefsky (1), for observing

at various depths the distribution of radioactive particles in a filter.

The experimental arrangement was designed to facilitate separation of the fiber bed into discrete layers for radioassay after exposure to the aerosol. We chose fibers with a high degree of uniformity in order to facilitate theoretical analysis. Raw dacron-polyester staple fibers (99 percent were  $11.3 \pm 0.8 \mu$ ) 3.8 cm long were separated into a uniform web by means of a carding machine, and were then rolled between sheets of fine paper to give a product which has a fiber-volume fraction of 0.28 and the appearance and feel of filter paper. A number of disks of this material, 3.8 cm in diameter and 0.04 cm thick, were placed in series between metal washers, enclosed in a cylindrical Teflon holder, and provided with O-ring seals to give a compact filtering device. For testing, a radioactive aerosol containing  $Zn^{65}$  was produced by using a Tesla coil to generate a spark between two pieces of neutron-irradiated zinc foil placed approximately 2 mm apart. A stream of air passed over the electrodes and carried the aerosol through the system containing the filters. An electron micrograph of samples of the aerosol collected on a membrane filter showed that it consisted of particles whose diameter ranged from 20 to 350 Å (median diameter, 120 Å; geometric standard deviation, 2.0).

Experiments were conducted over a wide range, the linear flow rates varying from 0.2 to 44 cm/sec. The data were analyzed by means of graphs (Fig. 1) in which the log of the relative radioactivity collected per layer was plotted against depth in the filter. Each layer contained approximately  $10^4$  cm of fiber per square centimeter of filter area, evaluated from disk weights and the density of the fiber. The abscissa in Fig. 1 has been simplified to show filter depth in centimeters, but the very small nonuniformity of the filter pads has been accounted for in plotting the data. In the region of low flow where Brownian diffusion is the dominant process for particle transport to the fiber, filtration efficiency decreased with increasing velocity. In the high-flow region, where inertial impaction is the principal process for particle transport, the efficiency increased with increasing velocity. In the intermediate-flow region, the interception range, where geometrical considerations of particle size and fiber size are very important, the filtration efficiency is largely independent of

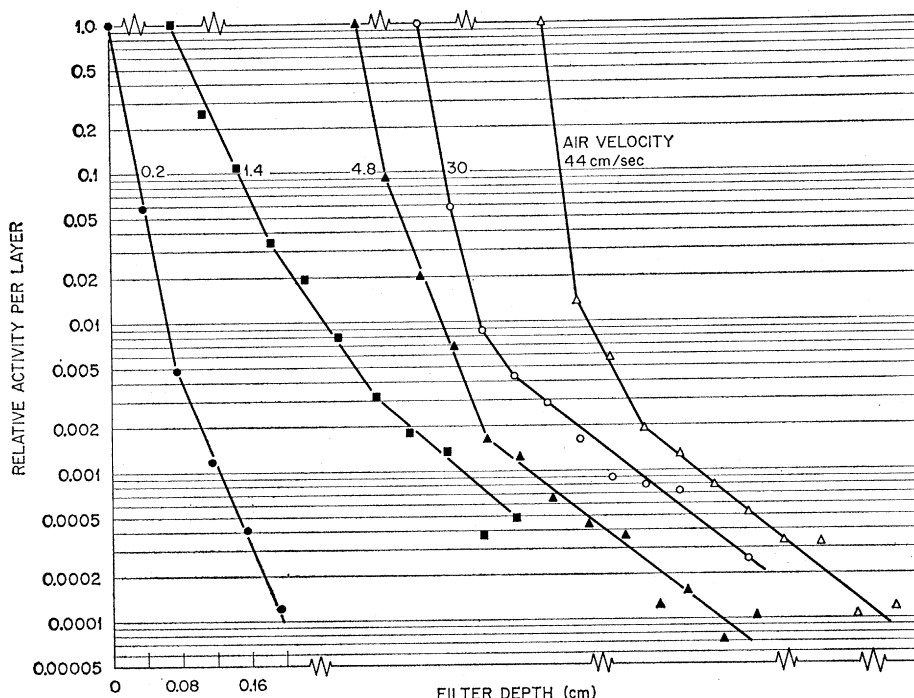


Fig. 1. Distribution of zinc activity at various depths in the filter; the diffusion regime at low air velocities can be recognized by the decrease of slope with increasing velocity.

velocity. Thus, the velocity dependence of the slope of these distribution curves identifies the mechanism of filtration, and the magnitude of the slope indicates the intensity of the filtering action.

The filtering action of fiber mat is the cumulative effect of particle removal by individual fibers. The collection efficiency of an individual fiber,  $\eta_a$ , is defined as the ratio of the cross-sectional area of the aerosol stream from which particles are removed to the projected area of the fiber in the direction of flow and may be obtained from the slope of the distribution curve on the semi-log graph. This slope may be substituted directly for  $(\ln N/N_0)/L$  in the expression derived by Langmuir (2) and confirmed by Davies (3) and Chen (4) for the single-fiber efficiency:

$$\eta_a = -\ln \frac{N}{N_0} \frac{\pi(1-\alpha)d_f}{4\alpha L} \quad (1)$$

where  $N_0$  and  $N$  are the upstream and downstream particle concentrations, respectively,  $\alpha$  is the volume fraction occupied by the fibers,  $L$  is the thickness of mat, and  $d_f$  is the fiber diameter, all in consistent units. Fiber efficiencies, evaluated for those experiments conducted at linear velocities of 0.2 to 2.5 cm/sec showed that  $\eta_a$  is proportional to  $1/\text{velocity}^x$ , where  $x$  is experimentally observed to be approximately  $1/2$ . This decrease of efficiency with increasing velocity indicates that, in this flow region, diffusion is the primary mechanism of filtration.

Using the method of Langmuir (2), which is based on diffusion of particles to the fiber surface in a fluid within a time equal to  $\pi d_f/2v$ , Stairmand (5) derived the following expression for the efficiency of diffusion collection:

$$\eta_a = \left( \frac{8D}{vd_f} \right)^{1/2}, \quad (2)$$

where  $v$  is the fluid velocity, and  $D$  is the diffusion coefficient calculated from the Einstein expression (6)

$$D = \frac{CkT}{3\pi\mu d_p} \quad (3)$$

Here  $d_p$  is the diameter of the particle,  $C$  is the Cunningham coefficient,  $\mu$  is the viscosity of the fluid, and the other symbols have their conventional meanings. The quantitative agreement between Eq. 2 and the experimental value of velocity dependence is confirmation that diffusion is the primary filtration mechanism under the specific conditions treated.

Particle diameters calculated from

the experimental data by Eq. 2 are shown in Table 1 for experiments at several different velocities. The double sets of figures represent the two groups of particle sizes demonstrated by the separate portions of the curves in the semi-log radioactivity-distribution graphs discussed. The use of Eq. 2 emphasizes the smaller end of the particle-size spectrum and the results agree well with the lower end of a particle distribution graph obtained from electron photomicroscopy of an aerosol prepared at 2.5 cm/sec. In the third column of Table 1 another set of diameters, weighted toward the larger particle sizes, was calculated as follows. Radioassay results from the filter mats, along with specific-activity data from the irradiated zinc-foil electrodes, were combined to give the total weight of zinc removed by the filters in each experiment. The number of particles produced was calculated from both the volume of air transported and a particle concentration for the aerosol (read from a condensation nuclei counter, Gardner Associates). The concentrations observed are consistent with those which would be expected from the geometry of the experiment and from the coagulation theory expounded by Whytlaw-Gray (7). From the weight of aerosol filtered (a density of 1 is assumed) (8) and the number of particles per experiment, an average diameter was calculated for the particles in each experiment. It should be noted that at the higher velocity, the particle diameters calculated theoretically are low compared with the radiochemical value, because the interception process is beginning to contribute to the filtration. The agreement between the three different methods for determining particle size is good.

The influence of mechanisms other than diffusion can be seen in the distribution curves for higher velocities (4.8 to 44 cm/sec) in Fig. 1. The velocity independence of the lower part of these curves identifies the interceptional mechanism. The slope of the upper part of these curves increases with air velocity indicating that inertial effects are important. Particles in the large end of the size range contribute greatly to this deposit because of their greater content of material.

We have demonstrated that the character of radioactive aerosols may be determined by an analysis of their filtration behavior in terms of diffusion, interception, and inertial impact by

Table 1. Comparison of particle sizes determined by different methods.

| Gas velocity<br>(cm/sec) | Particle diameter ( $\text{\AA}$ )<br>calculated by: |                       |
|--------------------------|--|-----------------------|
|                          | Diffusion<br>(Eq. 2)                                 | Radiochemical<br>data |
| 0.2                      | 104  | 180                   |
|                          | 168  |                       |
| 0.35                     | 100  | 160                   |
|                          | 180  |                       |
| 0.5                      | 52   | 135                   |
|                          | 98   |                       |
| 0.85                     | 36   | 105                   |
|                          | 70   |                       |
| 1.4                      | 35   | 80                    |
|                          | 105  |                       |
| 2.5*                     | 13   | 65                    |
|                          | 28   |                       |

\* An aerosol prepared at 2.5 cm/sec, examined by electron microscopy, had a median diameter of 120  $\text{\AA}$  and a geometric standard deviation of 2.0.

making use of observed depth distributions in specially prepared filters. In the diffusion region we have determined particle sizes from this behavior.

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#### Nitrous Oxide Produced by the Catalytic Oxidation of Organic Nitrogen Compounds

Abstract. Nitrous oxide is produced by the catalytic oxidation of organic nitrogen compounds of several distinct structural types with Hopcalite catalyst at a space velocity of 21,000 hours<sup>-1</sup> and at temperatures near 300°C.

The production of nitrous oxide in 70 percent yield by the catalytic oxidation of ammonia has been reported by Kobe and Hosman who employed a manganese oxide-bismuth oxide catalyst at 200°C (1). Krauss (2) and Nagel (3)