NOTES AND CORRESPONDENCE

Size and Nucleating Ability of AgI Particles

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Recently Gerber *et al.* (1970) showed experimentally how the ice nucleation behavior of an AgI smoke, generated by heating a small quantity of AgI-amine complex, depended on the size spectrum of the particles. Briefly, the size analysis of the active smoke particles entailed exposing the collecting foil of the Goetz aerosol centrifuge (Goetz *et al.*, 1960), on which the particles had been centrifugally deposited, to water saturation at a desired subfreezing temperature. During humidification with a slightly warmer ice surface placed several millimeters above the foil, the active AgI particles grew into discernible ice crystals. Since the coordinates of the foil are related to the Stokes diameter of the deposited particles, the locations of the crystals were used to determine the sizes of the corresponding active AgI particles.

When the size distributions of the active particles were compared to the electron microscope determined size distribution of all the generated particles, only a fraction of the total particles were observed to act as ice nuclei. Particles with projected diameters $\lesssim 200$ Å at -20C and $\lesssim 250$ Å at -15C were inactive, although the microscope analysis showed the peak in the size distribution to be ~ 150 Å.

A feasible explanation for these activity cutoffs was found in the theoretical work of Fletcher (1959a,b) who applied the Kelvin equation to determine the nucleation-inhibiting effects of surface curvature for small particles. When the experimental cutoffs were compared with Fletcher's predictions, good agreement was found for the appropriate case of nucleation by deposition at water saturation. It was therefore concluded that Fletcher's assumptions, in his classical derivation of that nucleation mechanism, were essentially correct.

The purpose of the present note is to revise the above conclusion in light of new experimental evidence. As a consequence of our participation in the International Workshop on Ice and Condensation Nuclei held at Ft. Collins in August 1970, it was discovered that the humidification technique used in the earlier work not only produced the desired water saturation over the centrifuge collecting foil, but it also appears to have produced a large inadvertent supersaturation caused by the introduction of warmer saturated air into the region of the foil as the ice surface was manually removed. Thus, only the nucleation before removal of the ice could have been due to deposition. Just after removal there was a large, visually observed increase in the particle activity probably due to nucleation by condensation-freezing. It is not necessarily proper to label the latter mechanism condensation-freezing, since the unambiguous use of the term freezing places the nuclei inside the water droplet. A better label, conceived by Edwards and Evans (1968), is the "puff" mechanism, where moist air is blown over the particles and causes water supersaturation. These workers, however, found no appreciable difference between the activation of AgI particles by the freezing and the "puff" mechanisms.

The earlier experiment was repeated with a similar AgI aerosol during the course of the Workshop, and again an ~ 200 Å cutoff was found for -20C. The cutoff for -16C was ~ 400 Å. When the supersaturation was avoided, and the particles were exposed only to water saturation, the activity cutoffs moved to larger sizes. Inactivity was found below ~ 500 Å for -20C and ~ 1500 Å for -16C.

Recently, Langer (private communication) also found an activity cutoff for 200 Å AgI particles at

−20C. He used the University of Minnesota's electrical particle analyzer (Whitby and Clark, 1965) to size all the particles and the acoustical ice nucleus counter (Langer, 1965) to simultaneously count the active particles. [The agreement between the present results for freezing and Langer's measurements with the acoustical counter, in which the nucleation by small AgI particles is now thought to be primarily as a result of contact between the particles and droplets at the temperatures discussed above (Langer, 1970; Middleton and Auer, 1970), suggests that contact nucleation might simply be a variation of the ordinary freezing behavior, as recently postulated by Fletcher (1970).]

The above cutoff data for freezing and deposition is for significantly larger particles than specified by Fletcher's classical nucleation theory. Much better agreement is found when the freezing data are compared to the modified nucleation-by-freezing theory of Fletcher (1969), which takes into account a statistical distribution of "active sites" on the surface of the AgI particles. When a nucleating activity of 1% is considered to be the cutoff, then the data for −20 and −16C approximately coincides with $\gamma = 1.0$ and $\beta = 10^{-4}$, which are, respectively, in Fletcher's notation, the width of the log-normal size distribution of the site areas and the fraction of the particle surface covered by sites. The cutoff for −15C agrees better with $\gamma = 0.8$ and $\beta = 10^{-3}$.

Although much more experimental evidence is necessary to evaluate Fletcher's present nucleation theory, it can be safely said that the effect of ice nuclei size on nuclei activity has been underestimated in the past.

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